



Carbonate and elemental accumulation rates in arid soils of mid-to-late Pleistocene outwash terraces, southeastern Wind River Range, Wyoming, USA

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Abstract: An important result of the past few decades of soil-geomorphic research is the awareness of the major role of dust and related carbonate precipitates in arid soils. The temporal evolution of soils developed on outwash terraces is still a matter of debate particularly regarding the type of evolution: progressive evolution and constant rates vs complex and/or interval-accumulation rates (progressive or regressive evolution possible). Soils on a chronosequence of outwash terraces just outside and downstream of canyon mouths of the Wind River Range (USA) were analysed for carbonate accumulation, organic carbon and poorly-crystalline pedogenetic oxyhydroxides stocks and pathways of chemical weathering. The soils and their carbonate stocks in the fine earth fraction (< 2 mm) can be used as a relative dating tool and appear to correlate with regional glacial chronostratigraphy [Pinedale (c. 20 ky), Bull Lake (with two different units around 160 and 260 ky) and Sacagawea Ridge (c. 660 ky)]. Carbonate stocks of soils on these terraces increase with increasing soil age, but at generally decreasing rates. A quasi-steady state situation, however, was not reached before 1 My. The time-split approach showed that CaCO_3 accumulation rates were the highest during marine isotope (MIS) cold stages (2.13 g $\text{CaCO}_3/\text{m}^2/\text{y}$), presumably due to a sparse vegetation cover during MIS2 (Pinedale) and 8 (lower Bull lake). Compared to organic carbon (c. 8–10 kg Corg/ m^2), the inorganic carbon stocks of the fine earth fraction were much higher (c. 60 kg Cinorg/ m^2). Although the dominant pedogenic process here appears to be carbonate accumulation, chemical weathering and elemental leaching also are significant. Weathering data were obtained using immobile elements and rare earth elements (REE) as tracers. Chemical weathering and related leaching were particularly notable for Na originating from plagioclase. In addition, pedogenetic Al and Mn oxyhydroxides accumulate (220 and 32 g/ m^2 , respectively, after 660 ky). These form at relatively low rates, however, when compared to moister and cooler conditions at higher altitudes in the Wind River Range. Due to the dry climate under which the terrace soils formed (annual precipitation 32 cm/yr–1), weakly crystalline and amorphous Fe-oxyhydroxide is rapidly transformed into crystalline forms (hematite) over time. Consequently, the stocks of weakly crystalline and amorphous Fe-oxyhydroxide was significantly lower in the 660 ky soils (50 g/ m^2) than in the Holocene soils (1000 g/ m^2). The development of soils on Quaternary outwash terraces in this region is controlled through the rates of aeolian influx over time as modified by glacial/interglacial climate variations rather than by moisture availability. Consequently, carbonate accumulation rates in these soils are ‘influx limited’ rather than ‘moisture limited’.

DOI: <https://doi.org/10.1016/j.chemgeo.2015.12.006>

Posted at the Zurich Open Repository and Archive, University of Zurich

ZORA URL: <https://doi.org/10.5167/uzh-135071>

Journal Article

Accepted Version



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Originally published at:

Dahms, Dennis; Egli, Markus (2016). Carbonate and elemental accumulation rates in arid soils of mid-to-late Pleistocene outwash terraces, southeastern Wind River Range, Wyoming, USA. *Chemical Geology*, 446:147-162.

DOI: <https://doi.org/10.1016/j.chemgeo.2015.12.006>

1 **Carbonate and elemental accumulation rates in arid soils of mid-to-late**
2 **Pleistocene outwash terraces, southeastern Wind River Range, Wyoming, USA**

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14 **Abstract**

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increasing soil age, but at generally decreasing rates. A quasi-steady state situation, however, was not reached before 1 My. The time-split approach showed that CaCO_3 accumulation rates were the highest during marine isotope (MIS) cold stages ($2.13 \text{ g CaCO}_3/\text{m}^2/\text{y}$), presumably due to a sparse vegetation cover during MIS2 (Pinedale) and 8 (lower Bull lake). Compared to organic carbon (c. $8 - 10 \text{ kg C}_{\text{org}}/\text{m}^2$), the inorganic carbon stocks of the fine earth fraction were much higher (c. $60 \text{ kg C}_{\text{inorg}}/\text{m}^2$). Although the dominant pedogenic process here appears to be carbonate accumulation, chemical weathering and elemental leaching also are significant. Weathering data were obtained using immobile elements and rare earth elements (REE) as tracers. Chemical weathering and related leaching were particularly notable for Na originating from plagioclase. In addition, pedogenetic Al and Mn oxyhydroxides accumulate (220 and $32 \text{ g}/\text{m}^2$, respectively, after 660 ky). These form at relatively low rates, however, when compared to moister and cooler conditions at higher altitudes in the Wind River Range. Due to the dry climate under which the terrace soils formed (annual precipitation $32 \text{ cm}/\text{yr}^{-1}$), weakly crystalline and amorphous Fe-oxyhydroxide is rapidly transformed into crystalline forms (hematite) over time. Consequently, the stocks of weakly crystalline and amorphous Fe-oxyhydroxide was significantly lower in the $\sim 660 \text{ ky}$ soils ($50 \text{ g}/\text{m}^2$) than in the Holocene soils ($1000 \text{ g}/\text{m}^2$).

The development of soils on Quaternary outwash terraces in this region are controlled through the rates of aeolian influx over time as modified by glacial/interglacial climate variations rather than by moisture availability. Consequently, carbonate accumulation rates in these soils are ‘influx limited’ rather than ‘moisture limited’.

Keywords: Chronosequence, arid soils, carbonate accumulation, weathering, Wind River Range

1. Introduction

50 Pleistocene climate cycles have driven multiple glacial-interglacial episodes in most middle-to-high
 51 latitude mountain regions of the world. In North America, the U.S. Rocky Mountains consist of
 52 numerous mountain ranges that contain records of multiple glacial episodes (e.g., Richmond 1986;
 53 Dahms 2004a). The Greater Yellowstone Ecosystem occupies a central role in our conception of
 54 Pleistocene glacial history since it has the type localities for the entire Pleistocene glacial
 55 succession identified in the U.S. Rocky mountains (see Richmond 1986; Dahms 2004a; Pierce
 56 2004). The Wind River Range (WRR) occupies the southern-most extent of the Yellowstone
 57 Ecosystem and was the locus of most early research into the Pleistocene glacial succession of the
 58 region (Blackwelder 1915; Richmond 1948, 1964, 1986; Mears, 1974). Most alpine valleys here
 59 were occupied by glaciers multiple times (Blackwelder 1915; Richmond 1973; Dahms 2004b).
 60 Morphostratigraphic evidence shows a minimum of four major glacial advances occurred here over
 61 the past ~1 My (Richmond, 1987; Hall and Shroba, 1995; Hall and Jaworowski, 1999; Dahms,
 62 2004b).
 63 An important result of the past few decades of soil–geomorphic research is the awareness of the
 64 major role of dust in arid (desert) and semi-arid regions of the world (e.g. Sauer et al., 2007; Muhs
 65 2013, Muhs et al. 2014). In the southwestern U.S. the influx of calcareous dust is central to the
 66 genesis of calcic and petrocalcic horizons in desert soils (Gile et al., 1981; Reheis et al., 1992;
 67 McFadden, 2013). Those soils that develop beneath desert pavements of the arid southwestern U.S.
 68 form in parent materials increasingly composed of entrapped dust that is subsequently translocated
 69 below the pavement to ‘build’ the soil from above (McFadden, 2013).
 70 Soils developed on Quaternary deposits of semi-arid (non-‘desert’) regions of the U.S. also exhibit
 71 significant contents of aeolian material (Colman and Pierce, 1986; Sorenson 1987; Reheis, 1988,
 72 1990; Muhs et al., 1990; Reheis, 1990; Dahms 1993; Dahms and Rawlins 1996) as do soils of
 73 alpine regions of the western U.S. (Marchand 1970; Birkeland 1973; Williams 1973; Thorn and

74 Darmody 1980; Boulding and Boulding 1981; Shroba and Birkeland 1983; Swanson 1985;
 75 Birkeland et al. 1987; Litaor, 1987, 1988; Dixon, 1992; Muhs and Benedict, 2006).
 76 We previously described soils developed on moraine chronosequences of the WRR and found clear
 77 aeolian signals in their particle size distributions, heavy mineral fractions and geochemical
 78 signatures (Dahms 1993, 2004b; Dahms and Rawlins 1996; Applegarth and Dahms 2001, 2004;
 79 Dahms et al., 2012). Few studies exist in this region, however, that describe soil chronosequences
 80 from the outwash terraces downstream of the moraine units. Soil characteristics are reported from
 81 terrace chronosequences of the Wind River in the central Wind River Basin upstream from the
 82 confluence of the Wind and Popo Agie rivers (Nettleton and Chadwick, 1991) and soils on the
 83 terraces of Rock Creek, a tributary of the Bighorn River are reported from the northern Big Horn
 84 Basin (Reheis, 1987, 1988, 1990). No previous data exists for tributary streams emanating from the
 85 glaciated valleys of the eastern slope of the Wind River Range.
 86 In this study we report a suite of newly-developed chemical weathering data for a chronosequence
 87 of soils on terraces of the three forks of the Popo Agie River near Lander, Wyoming. These data
 88 will contribute further to the development of soil chronosequences and will function as relative-age
 89 tools for correlating Pleistocene terraces in this region to their corresponding moraine units (Dahms,
 90 2010). In order to further develop evidence relating to the contribution of aeolian materials, and
 91 especially carbonate, to soil development over time on Quaternary alluvium in the Wind River-
 92 Yellowstone region (Hall, 1999), we describe a chronofunction for carbonate accumulation and try
 93 to derive, where possible, the corresponding interval accumulation rates (progressive or regressive
 94 evolution possible). Few data sets exist for long-term trends in production and accumulation of
 95 oxyhydroxides of Fe and Al in arid-lands soils such as these. Our results suggest that as weathering
 96 and pedogenesis proceeds over time, secondary mineral phases of Fe and Al form that initially have
 97 poorly/weakly crystalline characteristics, but develop strongly crystalline forms with time.
 98 Furthermore, disagreement exists as to whether such soils develop linearly and progressively or

99 if/when a quasi-steady state situation is ever reached (cf. Reheis, 1990; Reheis et al., 1992; Landi et
100 al., 2003).

101

102

103 **2. Study Region**

104 The study region is on the southeastern end of the Wind River Range, near Lander, Wyoming (Fig.
105 1). The drainages of the Popo Agie basin flow from the southernmost glaciated region of the range.
106 The Popo Agie basin drains most of the alpine region of the southeastern Wind River Range (Fig.
107 1). All three tributaries (the North, Middle, and Little Popo Agie) were occupied by Pleistocene
108 valley glaciers, resulting in the formation of a four-fold sequence of outwash terraces along each of
109 these streams as presumed floodplain aggradation during each glacial maximum gave way to stream
110 incision during the post-glacial period(s) (Vandenberghe, 2015).

111 The North Fork of the Popo Agie drains the Cirque of the Towers; the Middle Fork of the Popo
112 Agie drains Deep Lakes, Ice Lakes, Stough Lakes, and Leg Lake basins; the Little Popo Agie drains
113 the Silas and Christina Lake cirques (Fig. 1). The presence of well-preserved moraines near the
114 mouths of their canyons indicates that the valleys of all three of the rivers were occupied by glaciers
115 at least four times during the last ~1 Ma (Dahms 2004b and unpublished map data). We chose to
116 study the Popo Agie basin because the outwash terraces (Fig. 2) are well-expressed and appear
117 generally to correspond to previously-mapped terrace units along the Wind River (Chadwick et al.,
118 1997; Hancock et al., 1999; Dahms 2010) to which the Popo Agie river grades (Fig. 1) and on
119 which soils are previously described (Nettleton and Chadwick, 1991).

120 The Pleistocene terraces we studied are all within 5 miles of the mouth of their canyons (Fig. 2). All
121 terraces but the Holocene unit (PB-13-1) are strath terraces (e.g., Bierman and Montgomery, 2014)
122 formed as the rivers cut into the sedimentary units that crop out along the southeastern flank of the
123 WRR (Fig. 2). Terraces along the Little Popo Agie River have slightly different sedimentary and

124 soil parent material characteristics. These are inset into carbonate-bearing ‘Redbeds’ of the Red
125 Peak/Chugwater formations (Picard, 1993), so the coarse gravels of granitic and sedimentary origin
126 from the interior massif and inner canyon grade upward to clays, silts, and fine sand derived
127 exclusively from the redbeds (Fig 2d,e). Consequently, the parent materials of the soils on the Little
128 Popo Agie River terraces contain varying amounts of CaCO_3 (see Table 1). The soil parent
129 materials of the North and Middle Forks of the Popo Agie River are derived primarily from non-
130 calcareous granitic alluvium (Table 1) from the interior of the Wind River Range and from outwash
131 derived from upstream moraines (see Dahms, 2004a,b). The only terrace that contains fines derived
132 from redbeds in our present sequences along the North and Middle forks is the terrace represented
133 by sample GalPD (Fig. 2).

134 The climate of the lower slopes and piedmont of the eastern Wind River Range near Lander is
135 classified as semi-arid (Köppen *Bsk*) with a mean annual maximum temperature of 14°C , a mean
136 annual minimum temperature of 0°C and a MAT of 7°C (2015 US Climate Data). Mean annual
137 precipitation at Lander is 320 mm. All terrace sites of the study area are grassland/sagebrush steppe,
138 with riparian areas adjacent to the streams. The terrace soils (Fig. 3) are classified in the U.S.
139 taxonomic system as Aridisols and Inceptisols (Soil Survey Staff, 2014). The more well-developed
140 soils can be classified as Petrocryids/Calcicryids and the less-developed soils as Haplocryids.

141 Age control for terraces in this study is provided by geomorphologic mapping, including relative-
142 age relationships (Dahms unpublished maps), correlations with ^{10}Be and ^{26}Al boulder-surface
143 exposure ages from local moraine units of the Wind River Range (Gosse et al., 2003; Fabel et al.,
144 2004; Dahms, 2004b; Dahms and Egli, unpublished data) and correspondence to terraces and soils
145 of the Wind River (Hancock et al., 1999; Hall and Jaworowski, 1999; Jaworowski, 1992; Nettleton
146 and Chadwick, 1991). We also take into account the ages suggested for terraces along Rock Creek,
147 a tributary to the Bighorn River in the northern Bighorn Basin (Reheis, 1987). The Bighorn River is
148 the name assigned to the Wind River as it flows through the Bighorn Basin downstream of Wind

149 River Canyon (Fig. 1). For purposes of this study, we assign the following tentative ages to the
150 Popo Agie terraces and soils: Holocene ($5 \text{ ky} \pm 3 \text{ ky}$; profile PB-13-1), Pinedale ($20 \text{ ky} \pm 4 \text{ ky}$;
151 profiles GalPD and RCT-2), ‘lower’ Bull Lake ($160 \text{ ky} \pm 30 \text{ ky}$; profiles LWS, RCT-3, GR-13-1),
152 ‘upper’ Bull Lake ($260 \text{ ky} \pm 30 \text{ ky}$; profiles RCT-4, SB, GR-2) and Sacagawea Ridge ($660 \text{ ky} \pm 80$
153 ky ; profiles RCT-5, PB-13-2). We associate these ages, respectively, with Marine Isotope Stages 1
154 – 2 – 6 – 8 and 16. An additional numeric age to which we can correlate locally comes from a Lava
155 Creek ash deposit (c. 640 ky) reported by Jaworowski (1992) from the gravels of the highest
156 ‘airport’ terrace at Lander, from which no soil is reported here (Fig. 2B).

157

158

159 **3. Materials and methods**

160 *3.1. Sampling strategy*

161 Soils were sampled and described from hand-dug pits on flat summit-shoulder positions using
162 standard methods and horizon nomenclature (Fig. 3; Birkeland et al., 1991; Soil Survey Division
163 Staff 1993). To obtain a detailed and long-term chronosequence, we sampled 11 terraces having an
164 age of a few thousands of years (Holocene) to more than 600 ky. Profiles were dug to the C horizon
165 when possible. About 2 kg of soil material per horizon were taken for the analyses (Hitz et al.,
166 2002).

167

168 *3.1. Physical analyses*

169 Laboratory analyses of soil physical characteristics used standard techniques modified from Jackson
170 (1969), Klute (1986) and Singer and Janitsky (1986). Particle size-distribution of the $<2 \text{ mm}$
171 fraction was determined as weight percent (USDA scale) using the sieve-and-pipette method with
172 prior oxidation of organic matter by 30% H_2O_2 , removal of pedogenic carbonate via 0.5N HCL on
173 heat ($\sim 80^\circ\text{C}$) and is reported as weight percent (USDA grain-size scale). The bulk density (fine

174 earth and soil skeleton) was estimated from corresponding horizons of previously-reported soils for
175 which BD was determined from gravel-free paraffin-coated peds using an estimated gravel density
176 of 2.6g/cm^{-3} (Dahms, 2002, 2004).

177

178 3.2. Chemical analyses

179 Oven-dried samples ($70\text{ }^{\circ}\text{C}$) were sieved to $< 2\text{ mm}$ (fine earth) and homogenised with a sample
180 separator (Rentsch PT 1000). Soil-pH (0.01 M CaCl_2) was determined using a soil:solution ratio of
181 1:2.5. Carbonates were determined by using two different techniques. One consisted in the
182 dissolution of carbonates using HCl. The results were then compared with total Ca and Mg
183 concentrations obtained from XRF measurements. We performed an iterative speciation calculation
184 with the XRF data based on elemental contents and presented as oxides or carbonates (and fitted to
185 100% as total sum) to determine CaO , CaCO_3 , MgO and MgCO_3 species. Organic C and N contents
186 (determined in duplicate) of the soil were measured at $550\text{ }^{\circ}\text{C}$ using a C/H/N analyser (Leco). The
187 standard reference material is EDTA (Säntis Analytical, article no SA502092) having C = 41.09%,
188 H = 5.52%, N = 9.58%, O = 43.8% (measured values were C: $41.11\pm 0.08\%$; H: $5.53\pm 0.08\%$; N:
189 $9.57\pm 0.08\%$, O: $43.79\pm 0.08\%$).

190 Fe, Al and Mn concentrations were determined (in duplicate) after treatment with NH_4 -oxalate
191 (buffered at pH 3; McKeague et al., 1971). The extracts were centrifuged for 8 minutes at 4000 rpm
192 and filtered (mesh size $0.45\text{ }\mu\text{m}$, S&S, filter type 030/20). Element concentrations were measured
193 using atomic absorption spectroscopy (AAnalyst 700, Perkin Elmer). The blank values measured
194 (Fe: $-0.23 \pm 0.06\text{ mg/l}$; Al: $-0.04 \pm 0.8\text{ mg/l}$; Mn: $-0.005 \pm 0.004\text{ mg/l}$) were considered for the
195 elemental concentrations. Element concentrations were furthermore controlled using standard
196 addition (recovery $\geq 95\%$). The oxalate treatment extracts both the weakly- and poorly crystalline
197 phases and some of the organic phases, but normally does not dissolve the strong humus-metal
198 complexes (Mizota and van Reeuwijk, 1989). A portion of each oxalate extraction was analysed for

199 rare earth elements (REE) with an Agilent 8800 Triple Quadrupole (QQQ-ICP-MS). Prior to ICP-
200 MS analyses, standard solutions were prepared from SPEX multi-element plasma standard (Spex
201 CertiPrep, NJ, USA) at 0, 10, 50, 100, 500 and 1000 ppb to derive calibration curves. Concentration
202 of REEs were measured from standard solutions, reference material (CCRMP, REE-1), blank and
203 soil samples. Repeated measurements of the standards showed that the precision error was less than
204 8%.

205 The total elemental content was determined by X-ray fluorescence. Soil material was milled to < 63
206 μm in a tungsten carbide disc swing mill (Retsch® RS1, Germany). Powder samples (in duplicates)
207 of approximately 5 g material were analysed with an energy dispersive He-flushed X-ray
208 fluorescence spectrometer (ED-XRF, SPECTRO X-LAB 2000, SPECTRO Analytical Instruments,
209 Germany). The quality of the analyses was checked using a soil reference material (Reference Soil
210 Sample CCRMP SO-4, Canada Centre for Mineral and Energy Technology) with certified total
211 element concentrations. Element recovery and detection limits are given in Appendix A.

212

213 3.3. Soil mineralogy

214 We used XRD (X-ray diffraction) and DRIFT (Diffuse Reflectance Infrared Fourier Transform
215 spectroscopy) analyses to qualitatively describe the soils' mineralogical properties. An overview of
216 the mineral content in the fine silt and clay fraction (< 32 μm) was obtained by scanning randomly
217 oriented samples from 2 to 80°2 θ with steps of 0.02°2 θ at 10 s intervals using a Bruker AXS D8
218 Advance (CuK α).

219 DRIFT-spectra (Bruker, Tensor 27) were recorded from 4000 to 250 cm^{-1} using a powder
220 containing 30 mg of sample (10% of the total weight) and 270 mg KBr (90% of the total weight).
221 Prior to measurement, the samples were again oven-dried at 60° C. The FT-IR spectra were
222 interpreted using the OPUS 6.5 software.

223

224 3.4. Weathering indices

225 Weathering characterization is based on the calculation of elemental losses from soil or sediment
226 profiles. Relative elemental losses can be calculated using the open-system mass transport function

227 $\tau_{j,w}$ (Chadwick et al., 1990):

228

$$229 \tau_{j,w} = \left(\frac{C_{j,w} \cdot C_{i,p}}{C_{i,w} \cdot C_{j,p}} \right) - 1 \quad (1)$$

230

231 where i denotes the immobile element (Ti), $C_{j,p}$ (g/kg) is the concentration of element j in the
232 unweathered parent material and $C_{j,w}$ is the concentration of element j in the weathered product
233 (g/kg).

234 Mourier et al. (2008) showed that rare earth element (REE) geochemistry of lacustrine sediments
235 and soils is useful for reconstructing the history of landscape evolution. Normalised REE patterns
236 may provide a tracer of the degree of weathering of materials. In contrast to Mourier et al. (2008),
237 the REE concentrations were not normalised to a chondritic reference standard (Sun and
238 McDonough, 1989) but to the parent material of the soils to better facilitate the interpretation of
239 weathering behaviour (cf. Scarciglia et al., 2009, 2011) between sites. Normalisation against a
240 common reference helps to identify subtle fractionations and anomalies in elemental abundances
241 (Mourier et al., 2008). Only a portion of the REEs could be measured using XRF (La, Ce, Pr, Nd
242 and Sm). These elements were compared to the oxalate extractable fraction. In addition, the sample
243 composition (total concentration) was also compared to a set of major and trace elements of the
244 parent material. This set includes SiO₂, TiO₂, Al₂O₃, Fe₂O₃, CaO, MgO, Na₂O, K₂O, MnO, V, Cr,
245 Ni, Cu, Zn, Rb, Sr, Y, Zr, Nb, Ba, Pb, Th und U.

246

247 3.5. Stocks calculation and accumulation of pedogenetic products

248 Organic C and N stocks were calculated according to the following equation:

$$E_{stock,i} = \sum_{a=1}^n E_i \Delta z_i \rho_i (1 - RM) \quad (2)$$

where E_{stock} denotes the abundance (kg/m^2) of element i , E_i is the element concentration (kg/t), Δz_i the thickness of layer i (m), ρ_i = soil density (t/m^3) and RM the volumetric mass proportion of rock fragments.

To estimate the amount of pedogenically formed Fe, Al and Mn in soils, the concentrations of each compound in the soil column is compared to the parent material (or the layer with the least concentration). Using the concept of immobile elements, gains and losses of Fe, Al and Mn are derived using equation (3)

$$M_{j,w} = Me_{j,w} \left(\frac{\tau_{j,w}}{1 + \tau_{j,w}} \right) \Delta z_w \rho_w (1 - RM) \quad (3)$$

With n soil layers, the mass accumulation of the element j is given by:

$$M_{j,w}(\Delta z_w) = \sum_{a=1}^n Me_{j,w} \left(\frac{\tau_{j,w}}{1 + \tau_{j,w}} \right) \rho_w (1 - RM) \Delta z_w \quad (4)$$

where $\tau_{j,w}$ again corresponds to the mass transport function and Δz to the weathered equivalent of the columnar height.

The accumulation of inorganic C was calculated as the overall stock at the position where the parent material did not contain any carbonates. Soils on all the Little Popo Agie terraces (RCTs) and the GalPD terrace of the North Fork, however, contained small amounts of carbonate in the parent materials. In this case, accumulation (or losses) of inorganic C were calculated using the approach given in equations 3 and 4.

4. Results

4.1. General characteristics of the terrace soils

Except for profile RCT2, all investigated soils contain considerable contents of gravel that usually increases with increasing depths (Table 2). The soil texture is mostly clay loam, loam or silt loam.

273 In some cases (particularly C horizon), the texture can be characterised as sand, loamy sand or
274 sandy (clay) loam. Soil thickness (A+B horizon) varies between 40 and about 150 cm. Due to the
275 presence of carbonates the soils are all neutral or slightly alkaline (often between 7 and 8; Table 3).
276 Only the soils at PB-13-1 and GR-13-2 exhibit slightly acidic conditions in their upper horizons. In
277 several profiles, the B, BC and/or C horizons contain large accumulations of inorganic carbon. The
278 parent material at the RCT and GalPD sites contained some carbonates (13 – 22 g/kg inorg. C).
279 With increasing age of the terraces the average concentration of inorganic carbon increases in each
280 accumulation horizon (from a few g/kg to about 82 g/kg) and also the thickness of the carbonate-
281 rich subsoil horizons (Table 3; Figs. 3 and 4).
282 In contrast, the concentration of organic carbon shows no trend with age. The influence of organic
283 carbon on total C is manifested almost exclusively in the A horizons. With increasing soil depth the
284 overwhelming proportion of carbon exists in an inorganic form. The C_{org} content in A horizons is
285 not particularly high but is found to a considerable depth. Org. C/N ratios (Table 3) decrease with
286 increasing soil depth and exhibit low values in all profiles, particularly in the subsoils.
287 Pedogenic oxyhydroxides are present in all profiles. Fe(ox), Al(ox) and Mn(ox) usually decrease
288 with increasing soil depth. In surface soils, oxyhydroxides appear to be enriched passively due to
289 the accumulation of carbonates in the B, BC or C horizon, the weathering of silicate minerals, or
290 blown in as part of the aeolian influx..

291

292 *4.2. Rare earth elements and normalised elemental contents*

293 We used major and trace elements to assess pathways of chemical weathering and to assess aeolian
294 additions to the terrace soils. To compensate for the dilution effect of accumulated CaCO₃, the
295 considered elements were referred to Zr (Fig. 5). For several components (that are not related to
296 carbonates), the 20 ky terrace had the higher values and the 660 ky terrace soils (particularly in the
297 subsoil these differences are pronounced; Fig. 5). Although the terrace soils apparently have

298 received a considerable input of aeolian carbonates over time, some Mg, Ca and K has been lost due
299 to weathering of primary silicates. Mn is depleted in the subsoil but enriched in the A-horizon. In
300 addition, a substantial portion of Na has been leached in the subsoil. Most trace elements are
301 depleted but the signal is quite variable. Finally, Sr has accumulated with time in both A and B(k)
302 horizons. As Sr is strongly linked to the behaviour of Ca (and therefore carbonates) it indicates a
303 more or less continuous aeolian input and translocation of carbonates within the soils.

304 The values of the normalised REEs show that the oxalate-extractable fraction (and thus the fraction
305 found in the weathering products) is three to four orders of magnitude lower than the total REE
306 content. The normalized ratios (with respect to the total contents) are slightly lower in the B
307 horizons than in the A horizons. La, Ce and Nd are enriched over time in the A horizon but depleted
308 in the B(k) horizons. Similarly, the soils older than 20 ky show a slight accumulation of oxalate-
309 extractable REEs in the A horizon and a depletion in the B(k) horizons. This depletion (total and
310 oxalate-extractable fraction) probably is related to a dilution effect due to the accumulation of
311 carbonates in the subsoil.

312 All REE ratios are distinctly lower in the B(k) horizons when compared to the A horizons. As
313 byproducts of weathering, REEs are found to be incorporated into or adsorbed on secondary
314 minerals (clay minerals, pedogenic oxyhydroxides; Laveuf and Cornu, 2009). In the B horizons, the
315 decrease of oxalate-extractable La, Ce, Pr, Nd and Sm with time apparently is caused by dilution
316 owing to the accumulation of carbonates in the Bk horizon. Due to weathering of soil minerals, the
317 normalised oxalate-extractable REE fraction increases in A horizons with time (Fig. 6).

318 To study possible aeolian additions to soils, it is helpful to use chemically immobile elements. We
319 chose the elements Ti, Zr, Nb, Ce, and Y, all of which have a relatively high ionic potential and are
320 considered to be chemically immobile under most near-surface environments (Hutton, 1977; Taylor
321 and McLennan, 1985; Muhs and Benedict, 2006). According to Muhs and Benedict (2006), Tb and
322 Nb are found in ilmenite, rutile, anatase, titanomagnetite, sphene, and biotite. Zr is mostly present in

zircon, although Zr may also be found in other minerals (e.g. zirconolite). The REEs Ce and Y are related to a wide variety of minerals such as phyllosilicates (adsorption on e.g., micas, chlorite, other clay minerals), sphene, amphiboles and apatite (Muhs and Benedict, 2006). Ti/Zr, Ti/Nb and Ce/Y ratios measured in the A horizons, the horizons with carbonate accumulation (Bk, CBk, Ck) and the C horizons show a wide overlap (Fig. 7). The ratios demonstrate that the major chemical background of the various soil horizons does not vary appreciably and suggests that aeolian materials, which should be most concentrated in A and B horizons compared to C horizons, originate predominantly from local sources (e.g. vegetation free proglacial areas or sparsely vegetation-covered areas (Dahms, 1993; Dahms and Applegarth, 2001, 2004). Based on the above relations among the chemical and mineralogical soil properties, the Popo Agie terrace surfaces apparently have received significant amounts of dust that has led to, among other factors, an accumulation of pedogenic carbonate in the profiles over time.

335

4.3. Soil mineralogy

The primary minerals in the soil profiles are quartz, different types of feldspars, mica and calcite together with 1:1 (kaolinite) and 2:1 (vermiculite) phyllosilicates (Table 4); these mineral suites are consistent with the granitic nature of the local outwash material. 2:1:1 minerals are mostly absent. In addition, the parent materials for soils on the Little Popo Agie and the GalPd (North Fork) terraces contain calcite from the local redbeds. Hematite content also appears to increase with soil age (Table 4, Fig. 8).

343

4.4. Accumulation of inorganic and organic carbon and pedogenetically formed Fe_{ox} , Al_{ox} and Mn_{ox}

As noted above, the amount of inorganic carbon progressively increases in the soils with age (Fig. 9). This trend exhibits a logistic-curve form: during the first 20 ky the increase is moderate, a stronger increase is seen between 20 ky and 250 ky, and the ratio again slows for the older soils. At

the end of the sequence, a considerable amount of inorganic C is stored in the soil with average values of about 60 kg/m² in the fine earth fraction. With the exception of the 20 ky terrace soils (where a higher stock was measured with about 14 kg org. C/m²), the organic carbon stocks were in the range of 8 – 10 kg/m². In contrast to inorganic C, the organic carbon stocks did not change over time.

The pedogenic stocks of Fe_{ox}, Al_{ox} and Mn_{ox} all change considerably with time (Fig. 10) but the time-trends are not uniform. The Al_{ox} and Mn_{ox} abundances increase until c. 250 ky and then decrease to 660 ky. In contrast, the Fe_{ox} stocks in the soils steadily decrease with increased terrace age, with a marked loss of Fe_{ox} stocks between 5 and 20 ky. The stored amount at the start of the time sequence was about 1 kg/m²; by c. 600 ky the Fe_{ox} stocks decrease to only about 50 g/m².

5. Discussion

5.1. Carbonate accumulation

Gile et al. (1966, 1981) and Machette (1985) provide baseline data concerning the development of arid-lands soils emphasizing the systematic accumulation of pedogenic carbonate with time on chronosequences of geomorphic surfaces in the southwestern U.S. These authors showed that the calcium contained in pedogenic carbonate of arid-lands soils can derive directly from the parent materials, minerals present in the soil, or from atmospheric additions. As Muhs (2013) indicates, this type of soil development has been recognised in many desert regions of the world where the moisture balance is such that pedogenic carbonates are not fully leached from soil profiles but accumulate slowly, particularly in the subsoil. Where the geologic substrate provides little Ca, the majority of the Ca in pedogenic carbonate derives essentially from aeolian input (Gile et al., 1981; Muhs, 2013). According to McFadden (2013), virtually all calcium in pedogenic carbonate in the southwestern U.S. is derived from calcium-bearing dust that primarily accumulates in a very

373 common horizon of arid land soils – the calcic horizon.

374 The work of McFadden (2013), Reheis (1990; Reheis et al., 1992; Reheis and Kihl, 1995) and
375 Machette (1985) effectively demonstrates the regional importance of carbonate-rich aeolian influx
376 to geomorphic surfaces and the soils that develop in them. Soils reported from the Kyle Canyon
377 fans of Nevada (Reheis et al., 1992), the grassland and forests soils of Saskatchewan (Landi et al.,
378 2003) and various geomorphic surfaces of New Mexico and Utah (Machette, 1985) illustrate the
379 importance of regional carbonate accumulations in soils over time and allow us to generally
380 compare our results to soils developed in other arid regions of the U.S. More locally, Reheis' work
381 (1987) along Rock Creek in the northern Big Horn Basin allows us to compare our results with a
382 similar glacio-fluvial sequence in an adjacent area of the Middle Rocky Mountains.

383 The oldest soils of the Popo Agie River chronosequence are older than those in Nevada (where
384 stocks were available; the oldest Kyle Canyon soil was >800 ka) or Saskatchewan, yet our CaCO_3
385 stocks were mid-range between these regions (470 kg CaCO_3/m^2 compared to 630 kg/m² in the
386 Kyle Canyon fan of Nevada and 134 – 165 kg/m² in Saskatchewan).

387 Aeolian influx is the dominant mechanism for the carbonate accumulation over time in the soils
388 developed in the Popo Agie terraces. Calcium from atmospheric deposition enters the soil when it is
389 wetted and moves downward; calcium carbonate is precipitated on drying. Consequently,
390 carbonates tend to accumulate in the lower part of the zone that is wetted (Gile et al., 1981).

391 Reheis et al. (1992) reported that pedogenic CaCO_3 accumulated in the arid soils of southern
392 Nevada at a rate of 3 – 15 g/m²/y. Landi et al. (2003) derived CaCO_3 accumulation rates from the
393 grassland and forests soils of Saskatchewan in the range of 8.3 to 14.3 g/m²/y. In soils of the Rock
394 Creek terraces, Reheis (1987) reported CaCO_3 accumulation rates from 0.11 to 5.5 g/m²/y,
395 depending on distance from the range front. The values reported for southern Nevada and
396 Saskatchewan are clearly above those of the present study, but the accumulation rates for the Popo
397 Agie terrace chronosequence (0.19 - 2.13 g/m²/y using an interval accumulation approach and 0.71

398 – 1.96 g/m²/y using a linear approach; Table 5) are within the range of those reported for the Rock
 399 Creek terraces, which lie in a similar setting and climatic regime. In Table 5, interval accumulation
 400 rates of carbonate and accumulation rates using a ‘linear approach’ (by dividing the carbonate
 401 stocks stored in the soils by the corresponding surface age) are presented. The temporal evolution of
 402 the inorganic C abundance in the soils is strongly non-linear (Fig. 9). In soil chronosequences,
 403 concentrations or stocks of a component are compared to surface age. The average process rate is
 404 often calculated by dividing the measured stocks or concentrations by the soil age (Schlesinger,
 405 1990) – a procedure that in many cases does not reproduce the correct values (because a linear trend
 406 is implicitly assumed). A time-split approach or the derivation of a regression curve (having in fact
 407 an infinite number of time-splits) would be more precise.

408 We calculate only a net accumulation rate of CaCO₃. The present-day deposition of dust is probably
 409 higher and consists of more than just carbonates, but we assume that part of it is leached from the
 410 profiles or removed by wind erosion (Hall, 1999; Applegarth and Dahms, 2001). A two-year record
 411 of aeolian influx rates in alpine areas of the Wind River Range yielded values of 2.5 – 3.3 g/m²/y
 412 for mineral dust (Dahms and Rawlins, 1996) and Ferrier et al. (2011) estimated even higher long-
 413 term dust fluxes (c. 3 to 13 g/m²/y) for the western U.S.

414 Our results suggest that carbonate accumulation, and by extrapolation dust deposition, has varied
 415 over major glacial-interglacial cycles. Carbonate stocks of the Popo Agie soils are the lowest in the
 416 younger soils and the highest in the older soils, but the stocks do not increase uniformly over time
 417 (Table 5). For example, the average CaCO₃ accumulation rate during the 260 to 660 ky period was
 418 the lowest (0.19 g/m²/y; using a linear approach, then the average value between 0 – 660 ky would
 419 be 0.71 g/m²/y; Table 5). The accumulation rate increased to 0.79 g/m²/y during the 260 – 160 kyr
 420 interval, then increased again to 2.13 g/m²/y from 160 – 20 ky (for the period 0 – 660 ky, the linear
 421 approach gives 1.96 g/m²/y; Table 5), after which the accumulation rate decreased to 0.79 g/m²/y in
 422 the post-glacial (Pinedale – Holocene). Independent of the calculation procedure (interval

423 accumulation or linear approach), highest rates are related to the period 20 – 160 ky BP.

424 A moderately comparable pattern of carbonate accumulation is found in the northern Big Horn
425 Basin (Reheis, 1987). The lowest accumulation rate ($0.11 - 1.5 \text{ g/m}^2/\text{y}$) is reported in soils of the
426 oldest terraces ($>400 \text{ ky}$) and the highest rates ($1.0 - 5.5 \text{ g/m}^2/\text{y}$) are estimated from the youngest
427 soils. Machette (1985) reported a similar general pattern of CaCO_3 accumulation (older = low;
428 younger = high) from New Mexico and Utah.

429 Carbonate stocks in the Popo Agie terrace soils continue to increase with time but do not appear to
430 reach a quasi-steady state before about 1 My. Reheis (1990) also observed that arid soils develop at
431 linear rates over several 100 ky. In this case soil development reflects progressive dust addition that
432 may be little affected by leaching or erosion. We expect basin soils to accumulate large amounts of
433 Mg and Ca because the climate is dry enough for these soils to retain much of the carbonate derived
434 from aeolian dust (Reheis, 1990).

435

436 5.2. Chemical weathering pathways

437 Chemical weathering and leaching of elements are both related to a decreasing value of the open-
438 system mass transport function $\tau_{j,w}$ (negative values represent losses and positive values gains). As
439 we expected, this is in general not the case for Ca, because it is accumulated during pedogenesis.
440 The higher the carbonate content in the soil is, the higher is the corresponding τ_{Ca} (Fig. 11).
441 However, some weathering and leaching of elements do take place. With increasing soil age the
442 carbonate content and τ_{Ca} increase, but the τ_{Na} decreases (Fig. 11). Na predominantly derives from
443 plagioclase and our τ_{Na} values demonstrate that plagioclase is strongly weathered in the older soils -
444 a finding that is also supported by the normalised data of CaO and Na_2O to the parent material (Fig.
445 5). In the Wind River Range, plagioclase is transformed over time into kaolinite, illite and smectite
446 (Mavris et al., 2015). With increasing weathering state, also the REE contents (total and oxalate-
447 extractable content) were mostly higher in the A horizons. Chemical weathering accounts for the

largest losses of REEs from the parent material (Laveuf and Cornu, 2009). According to Scarciglia et al. (2009, 2011), weathering of primary minerals and pedogenetic processes give rise to REE adsorption onto reactive sites of organic matter and clay particles. Clay eluviation and illuviation furthermore contribute to the elemental fractionation (Laveuf and Cornu, 2009; Scarciglia et al., 2009, 2011).

Under relatively moist conditions, the amounts of pedogenetic, weakly crystalline or amorphous oxyhydroxides increase with time (e.g., Egli et al., 2001; Howard et al., 2012). A similar trend was detected in the wetter, cooler high altitude subalpine-to-alpine environments of the Wind River Range (Dahms et al., 2012). With progressing soil evolution portions of the amorphous forms may be transformed into goethite (FeOOH ; cf., Ellis and Atherton, 2003). In sharp contrast, the abundance of pedogenetically formed Fe_{ox} (weakly crystalline and amorphous forms) in the Popo Agie terrace soils decreased strongly with increasing time (Fig. 9). Some of the weakly crystalline Fe may also derive from aeolian input – its contribution, however, does not seem to be that high because no clear Fe accumulation with time was detected (Fig. 5). We suggest that, due to the semi-arid climate, a relatively fast transformation of weakly crystalline and amorphous Fe-forms into crystalline Fe-oxides or -hydroxides (that are not dissolved using oxalate) is likely. This idea is supported by the XRD results: more hematite (Fe_2O_3) is detected in the older soils (Table 4; Fig. 8) whereas goethite seems to be nearly absent (presumably due to the dry climatic conditions). This weathering relationship is further supported by Quinton et al. (2011) who demonstrated that the absolute and relative hematite abundance in the Little Popo Agie soils (profiles RCT-2-to-5) increased with soil age. Aeolian influx of crystalline Fe-minerals does not seem to be a major process contributing to such a change, because no distinct accumulation (nor depletion) of Fe was detected (Fig. 5).

The pedogenetically-formed Fe stocks of the Holocene soil (PB-13-1) corresponds to what has been measured at alpine moraine sites of the Wind River Range (Dahms et al., 2012). At cooler and

473 moister sites (montane-to-alpine) the formation of Fe_{ox} continues over time, whereas the
474 transformation of weakly- and poorly-crystalline phases into crystalline forms is enhanced at the
475 terrace sites.

476 Stocks of pedogenic Al-forms in the Popo Agie soils tend to increase with time. This is an expected
477 result of the weathering process; however, the stocks remain at a rather low level even after several
478 100 ky (200-300 g/m²) suggesting modest weathering compared to alpine moraine soils of the WRR
479 (Dahms et al., 2012).

480 Gile et al. (1981) observed that in arid environments the amount of organic carbon generally
481 increases with the clay content. Our observations show that the relation between organic carbon and
482 clay and silt stocks is rather weak (Fig. 12). We see a higher correspondence, however, between
483 nitrogen and silt. We do not really understand why, but we hypothesise that larger amounts of
484 highly-charged phyllosilicates are available in the silt fraction that enable better adsorption of
485 organic matter (having proteinaceous compounds) and protection from microbial decay. The higher
486 amount of highly-charged phyllosilicates could be related to silt-sized mineral grains (e.g.,
487 plagioclase, feldspar and micas) that are weathered in situ to clay minerals. Kleber et al. (2007),
488 Kögel-Knabner et al. (2008) and others show the importance of the mineral fraction in OM
489 protection. According to Kleber et al.'s model the formation of particularly strong organo-mineral
490 associations is favoured in the so-called 'contact zone' in situations where proteinaceous materials
491 unfold upon adsorption and thus increase the adhesive strength by adding hydrophobic interactions
492 to electrostatic binding. This could be a reason why such low C/N ratios have been measured in our
493 soils. In addition, more aggregates simply may be formed by silt that physically protects organic
494 compounds from decay. Another explanation for the very low C/N ratio particularly in the deeper
495 soil horizons, might be due to the exceptionally high accumulation of nitrate beneath the active
496 rooting zone (Graham et al., 2008; Menon et al, 2010). According to these authors, soil resources
497 (water and organic carbon), precipitation, net dryfall and rare leaching events are the primary

498 factors that lead to accumulation of NO_3^- -N particularly in this region. Previous measurements of
499 N-deposition in the WRR (Galbraith et al., 1991) showed that a considerable portion of NO_3^- -N is
500 imported from upwind sources (presumably) west of the WRR.

501

502 *5.3. Climatic inferences*

503 Reheis (1987) observed that the ‘Basin Soils’ located farthest from the range front preserve the
504 most complete record of climate change along the Rock Creek chronosequence. The use of
505 alternating clay cutans and CaCO_3 layering from thin-section analyses enables Reheis to infer
506 alternations of glacial/interglacial climates in these soils. The inference is that climate fluctuations
507 over time here were enough to cause these changes, but not enough to destroy previously-developed
508 soil characteristics – hence the continuous accumulation of carbonate (regardless of rate) over time.
509 Such features have often a polygenetic origin. Clay increase and cause of clay orientation is often
510 not due to a single genetic process or event (Gunal and Ransom, 2006).

511 The Popo Agie River soils occur in a similar environment, and so most likely also reflect climate
512 fluctuations over the past c. 700 kyrs. We did not thin-section our soils and so do not have data
513 similar to those of Reheis (1987). Reheis concluded that it is likely that the variable depths of clay
514 and carbonate in the older Rock Creek terraces soils were most likely a result of increases in
515 wetting depth due to an increase in available water capacity (AWC) coincident with the addition of
516 aeolian fines over time rather than to changes in overall climate variability (e.g., higher
517 precipitation levels during interglacial periods). Overall, the Rock Creek soils data suggest an
518 overall pattern of less precipitation during glacial periods. Alternately, Nettleton and Chadwick
519 (1991) suggested that moisture conditions during the Pleistocene in the lower Wind River Basin
520 near Riverton were not appreciably different from those at present and also suggest that dust
521 deposition rates have not appreciably changed.

522 Our CaCO_3 accumulation data are similar to those for Rock Creek and it seems reasonable that
523 some carbonates in the soils would be dissolved during periods of greater moisture. However, our
524 results suggest that CaCO_3 accumulates most effectively in the soils of the western Wind River
525 Basin during the cool, windy glacial periods rather than during warmer, less windy interglacials
526 (Table 5). Hall and Jaworowski (1999) have suggested that A horizons of Bull Lake-age are thinner
527 and Bt horizons are closer to the surface than on soils of nearby Pinedale moraines due to erosion
528 caused by the cold foehn winds off the local valley glaciers. Studies of Greenland and the Antarctic
529 ice cores show that MIS glacial stages are drier and windier (and thus dustier) than interglacials
530 (e.g., Fuhrer et al., 1999; Svensson et al., 2000). Thus, it seems reasonable to assume that more Ca-
531 rich materials normally are in flux in a glaciated region during time periods that are predominately
532 glacial than during warmer periods. It appears that, on average, our soil CaCO_3 accumulation rates
533 (and associated aeolian deposition rates) were the highest during MIS 6-2 (c. 160 ky; Cohen and
534 Gibbard, 2011). Note that the time-span 160-to-20 ky predominantly covers cold MIS phases. The
535 other time-split periods (660-to-260 ky and 260-to-160 ky) include a comparatively larger
536 proportion of warm (interglacial) phases. Machette's (1985) model for long-term carbonate
537 accumulation shows that soils in locations with climatic regimes similar to that of the Popo Agie
538 (Buena Vista & Boulder/Denver, Colorado) fall into the 'influx limited' region of the model
539 (Machette, 1985). This model suggests that the variations observed in the Rock Creek, Wind River,
540 and Popo Agie soils are more likely to be caused by variations in aeolian influx under the influence
541 of climate variability than either influx rates or glacial/interglacial climate variations acting alone.
542 This reasoning should include a decrease in vegetation cover during glacial periods. We suggest
543 that change in rates of soil carbonate accumulation does not exclusively indicate a climate signal
544 but probably is overprinted by limits to the supply of sediments, the availability of aeolian material,
545 balanced by the effect(s) of increasing loss by erosion of fines as the surfaces age and the
546 decreasing ability of the surface soil to accumulate more eolian fines as pore spaces fill with

547 infiltrated silt and clay. These factors may well influence the (apparently) decreasing accumulation
548 rates detected in the older soils.

549

550

551 **6. Conclusions**

552 Chronosequences are a fundamental tool for studying and representing change in Earth surface
553 systems. Increasingly, chronosequences are understood to be much more complex than a simple
554 monotonic progression from a starting point to a stable end-state (Phillips, 2015). In this case, the
555 soil physical and chemical characteristics combined with the presumed age associations allow us to
556 derive interval accumulation rates of CaCO_3 that appear to show varying rates of dust deposition
557 over several glacial-interglacial cycles. The development trajectories thus show a certain sensitivity
558 to disturbances and changes (Phillips, 2015).

559 The well-preserved terraces along the Popo Agie River of the southeastern Wind River Range
560 allows us to develop a relative-age chronosequence for soil development on (mostly) granitic
561 alluvium and to compare soil development here with soils on similar terrace sequences elsewhere in
562 the western U.S. Carbonate stocks in the fine earth of the soils increase with increasing soil age and
563 are thought to be related to aeolian influx over time and influenced by the major cycles of mid-to-
564 late Pleistocene climate change represented in our chronosequence [Holocene, Pinedale (20 ky),
565 early and late Bull lake (160 and 260 ky) and Sacagawea Ridge (c. 660 ky)]. By inference of the
566 carbonate stocks, aeolian influx of inorganic carbon apparently was lowest in this area during the
567 period of c. 260 – 660 ky and the highest during the period of c. 160 – 20 ky.

568 A quasi-steady state situation (where inputs of materials to the soil system and outputs via erosion
569 and weathering are almost equal) does not appear to have been reached by 660 ky. The change in
570 carbonate accumulation rates in the soils may exclusively indicate a climate signal but is probably

571 also related to a supply limitation of sediments, the subsequent disposability of aeolian material,
572 and increasing erosion of older soil surfaces.

573 We suggest a scenario where the long-term carbonate accumulation rates in soils of Quaternary
574 glacial, alluvial, and glacio-fluvial sequences of this region are controlled through the rates of
575 aeolian influx over time as modified by glacial/interglacial climate variations rather than by
576 glacial/interglacial variations in moisture availability. Thus we consider the carbonate accumulation
577 rates in these soils to be 'influx limited' rather than 'moisture limited' (Machette, 1985) so that the
578 *soil-forming regime* of these soils is more closely related to that of the semi-arid Rocky Mountain-
579 Intermountain region than to the arid southwest of New Mexico, Arizona, and southern Nevada and
580 California.

581 Compared to other published results, the carbonate accumulation rates determined for the Wind
582 River Range terraces were relatively low. Despite the dominance of carbonate accumulation as a
583 pedogenetic process, chemical weathering and leaching of primary minerals occurred (e.g.
584 plagioclase weathering coupled with a subsequent Na release). In addition, pedogenetic Al and Mn
585 have formed, but at relatively low rates when compared to moister and cooler conditions in the
586 Wind River Range. Due to the dry climate, weakly crystalline and amorphous Fe-forms seemed to
587 be transformed relatively quickly into crystalline forms (hematite). With increasing age, inorganic
588 carbon (with an average at the oldest sites of 60 kg C_{inorg}/m²) becomes the lion's share of the stored
589 total carbon. Organic carbon remained relatively at a stable level (about 8 – 10 kg C_{org}/m²) over the
590 entire soil formation phase.

591

592

593 **7. Acknowledgements**

594 We would like to thank S. Röthlisberger and M. Kovacic for their help in the laboratory at the
595 University of Zürich. Students in the Environmental Sediment Analysis class (Geog4220) at the

596 University of Northern Iowa helped Dahms to develop the data for % CaCO₃. We are, furthermore,
597 indebted to Marith C. Reheis and an unknown reviewer for their helpful comments on an earlier
598 version of the manuscript.

599

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Figure captions

Fig. 1. Location of the Popo Agie Basin on the southeastern flank of the Wind River Range in west-central Wyoming. Note the three forks of the Popo Agie grade to the Wind River south of Riverton in the central Wind River Basin and that the Wind River becomes the Big Horn River at the northern mouth of Wind River Canyon (located just north of the lake at the mid-upper right of the image (Google Earth Pro).

Fig. 2. Location of the studied terrace elements and associated soil profiles along each of the three forks of the Popo Agie River: A) the North Fork, B) the Middle Fork (together with the airport terrace AT (Jaworowski, 1992); see text), C) the Little Popo Agie (Red Canyon), D) terraces of RCT2-5 in Little Popo Agie and E) view toward the north from RCT-4 (Little Popo Agie). Photos generally are SE-to-NW (for A) to C)). The terrace/soil locations are within 2.3 miles of the mouth of North Fork Canyon, 5.0 miles from the mouth of Sinks Canyon (Middle Popo Agie) and within 0.5 mile of the mouth of Little Popo Agie Canyon.

Fig. 3. Investigated soil profiles with designated horizons. For RCT5, the inset shows the character of the Bkm (cemented ‘petrocalcic’) horizon of the U.S. Soil Taxonomy.

Fig. 4. A) Average concentration of inorganic and B) total carbon as a function of soil depth and age. Values are averages of the soils within the corresponding age classes.

Fig. 5. Spider diagram of normalised (to the parent material = PM) A) major elements in the A horizon, B) trace elements in the A horizon, C) major elements in the B(k) horizon and D) trace elements in the B(k) horizon. The concentrations are given as a function of surface age.

To account for the dilution effect of CaCO_3 , the contents were ratioed to the immobile element Zr:

$$\text{Conc. measured(norm)}/\text{Conc. PM (norm)} = \frac{\frac{C_s}{Zr_s}}{\frac{C_{PM}}{Zr_{PM}}} \quad \text{where } C_s = \text{element concentration of the sample, } Zr_s = \text{Zr concentration of the sample, } C_{PM} = \text{element concentration of PM (parent material) and } Zr_{PM} = \text{Zr concentration of PM.}$$

the sample, Zr_s = Zr concentration of the sample, C_{PM} = element concentration of PM (parent material) and Zr_{PM} = Zr concentration of PM.

Fig. 6. Normalised (to the parent material = PM) REE concentrations for some selected profiles as a function of terrace age. The total content and the oxalate-extractable fraction of selected REEs are shown for A) the A-horizon and B) the B(k)-horizon. The content of Sm in the A horizon was below the XRF detection limit.

Fig. 7. Binary diagram of concentration ratios of A) Ti/Zr vs Ti/Nb and B) Ti/Zr vs Ce/Y for samples of the topsoil, the horizons with carbonate accumulation (Bk, CBk, Ck) and the C horizon from the various sites.

Fig. 8. X-ray diffraction pattern for three samples from the soils PB-13-2 (Bw horizon), GR-13-1 (Btk) and PB-13-1 (AB horizon) having different ages. The diffraction peaks associated with quartz (Q), calcite (Cc) and hematite (H) are labelled.

Fig. 9. A) Inorganic and B) organic carbon stocks (average \pm standard deviation) as a function of age (\pm estimated error range).

Fig. 10. Average (\pm SD) accumulation (abundance) of pedogenetically formed A) Fe, B) Al and C) Mn in the entire profiles as a function of terrace age (\pm error range).

Fig. 11. A) Comparison of the relative losses or gains (open system mass transport function τ) of Ca with the carbonate content and B) relative mass changes of Ca vs. Na.

Fig. 12. Comparison of organic C stocks with the abundance of A) clay and B) silt of the individual soil horizons. Significant correlations only exist between C) N stocks and clay abundance and D) N and silt abundance.

Figure 1
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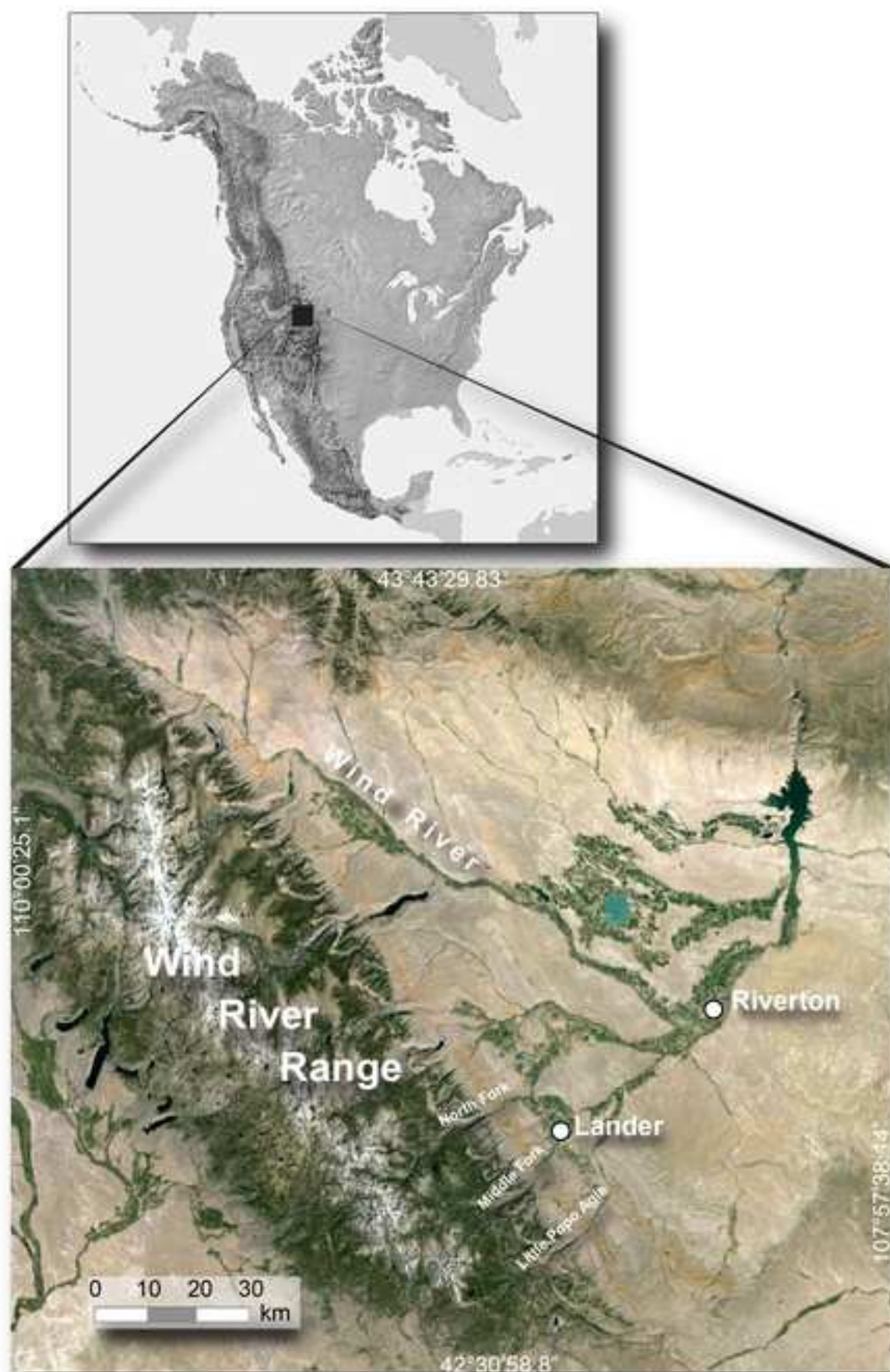


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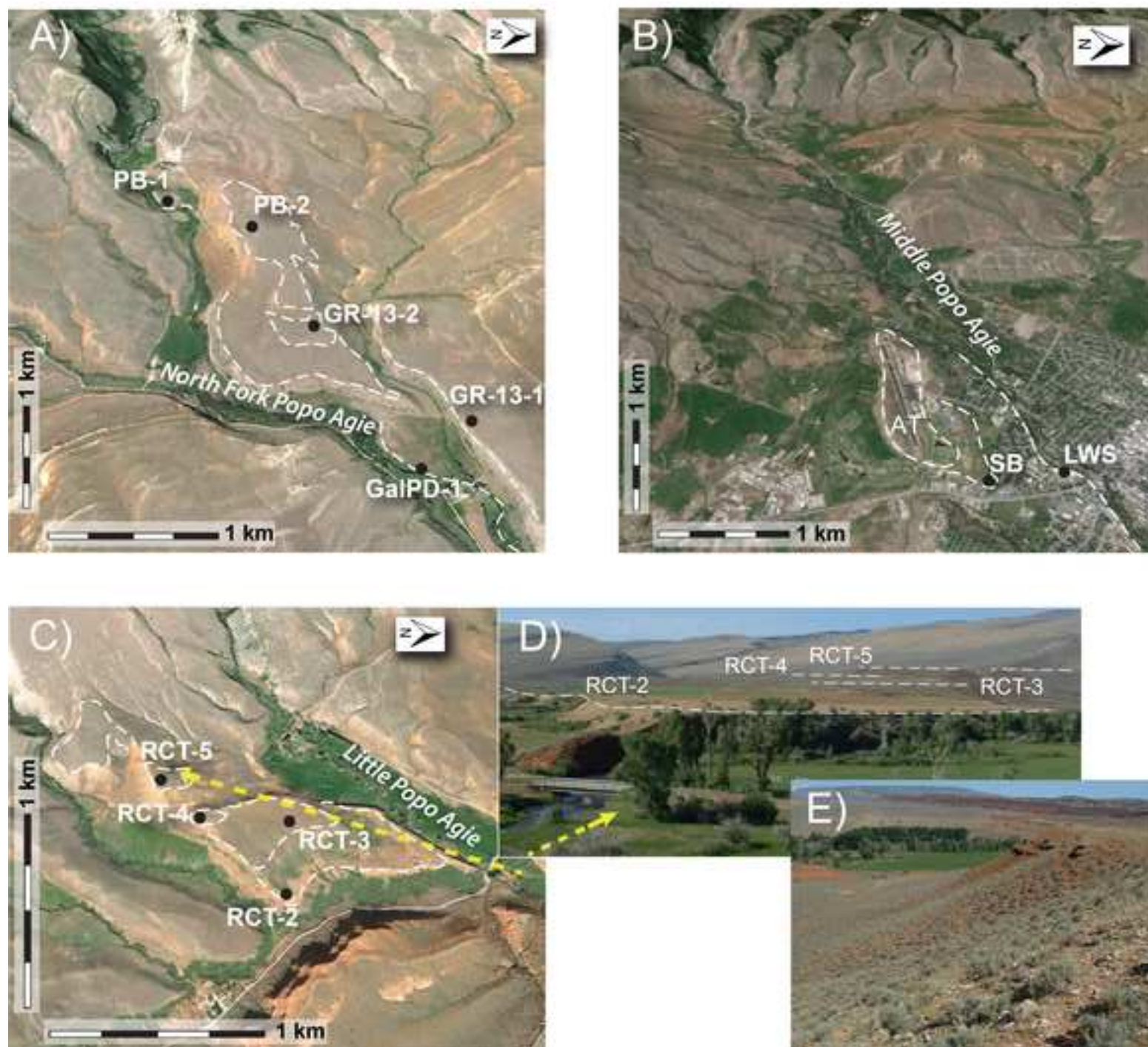


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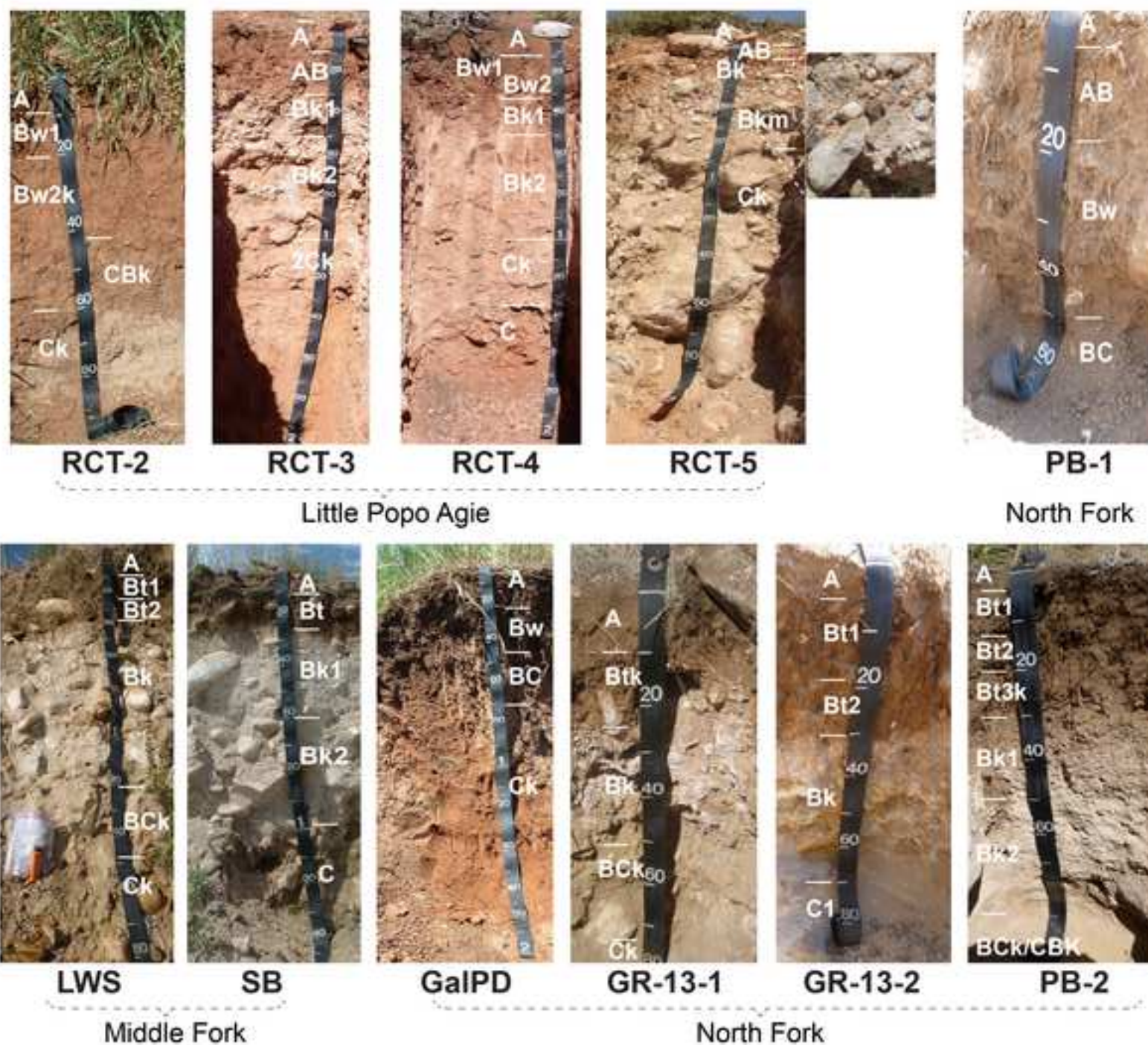


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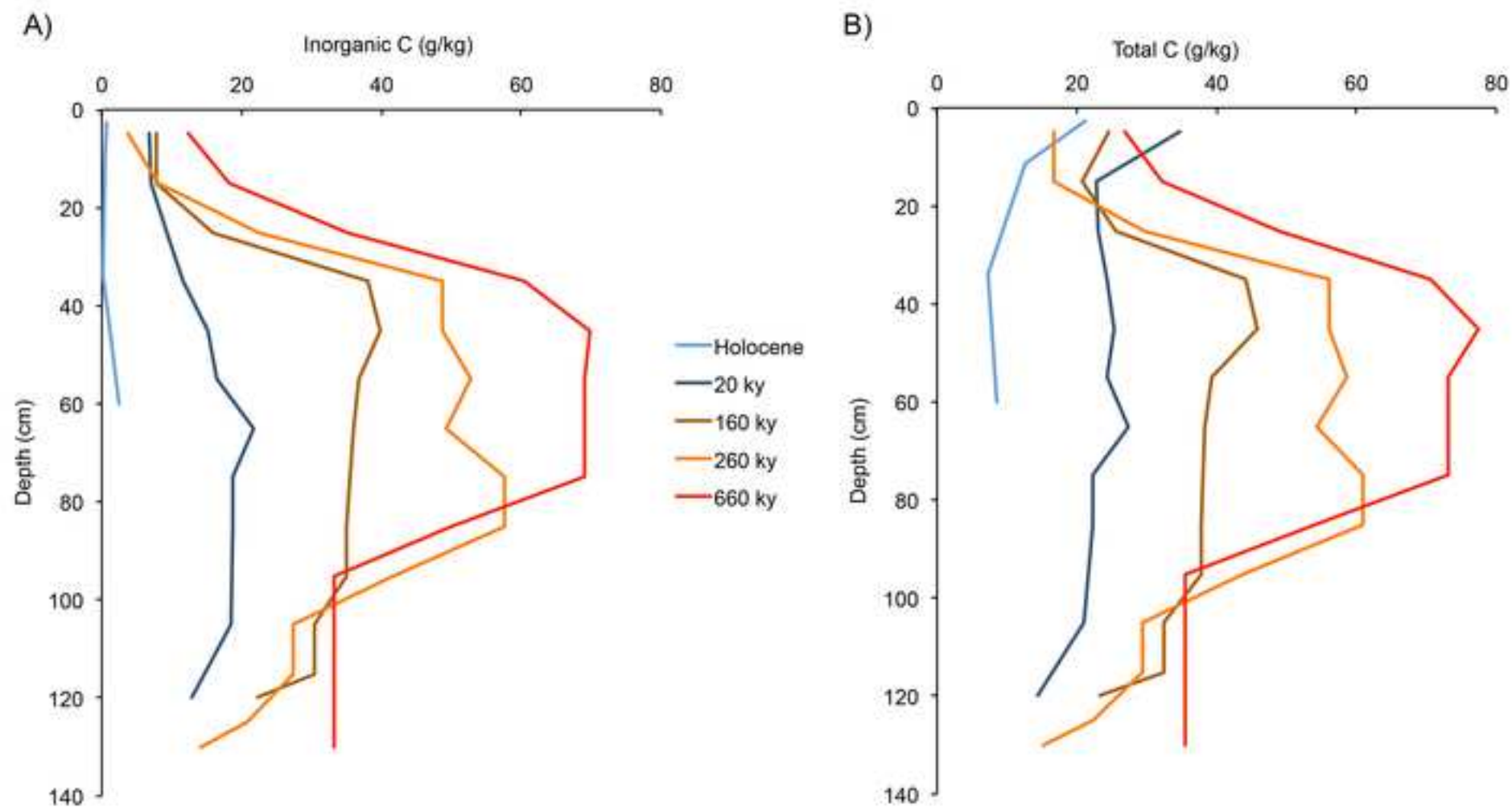


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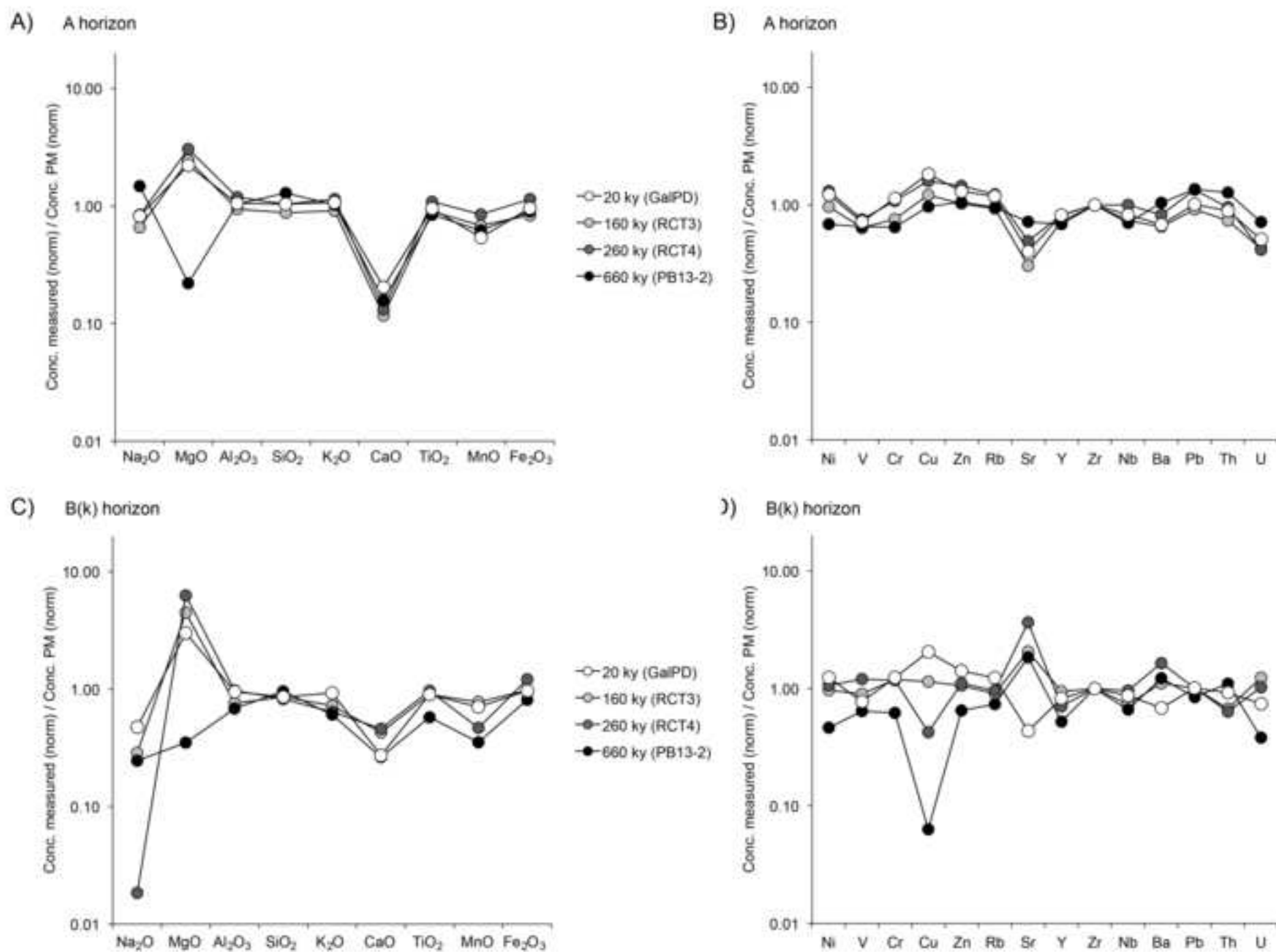
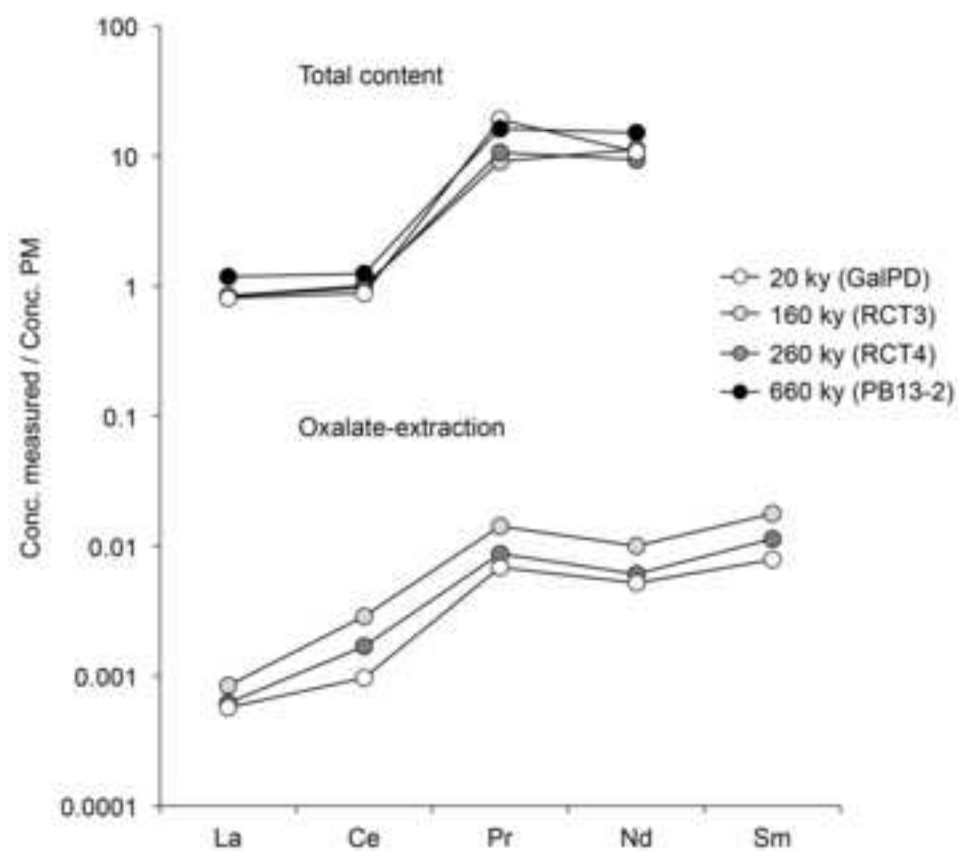


Figure 6
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A) A-horizon



B) B(k)-horizon

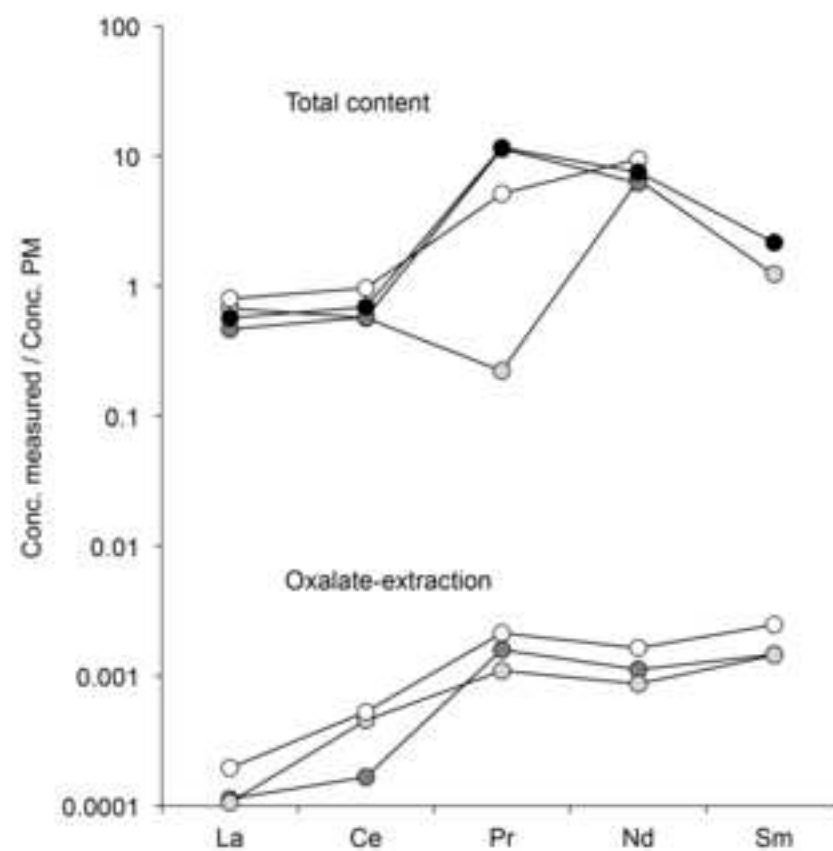


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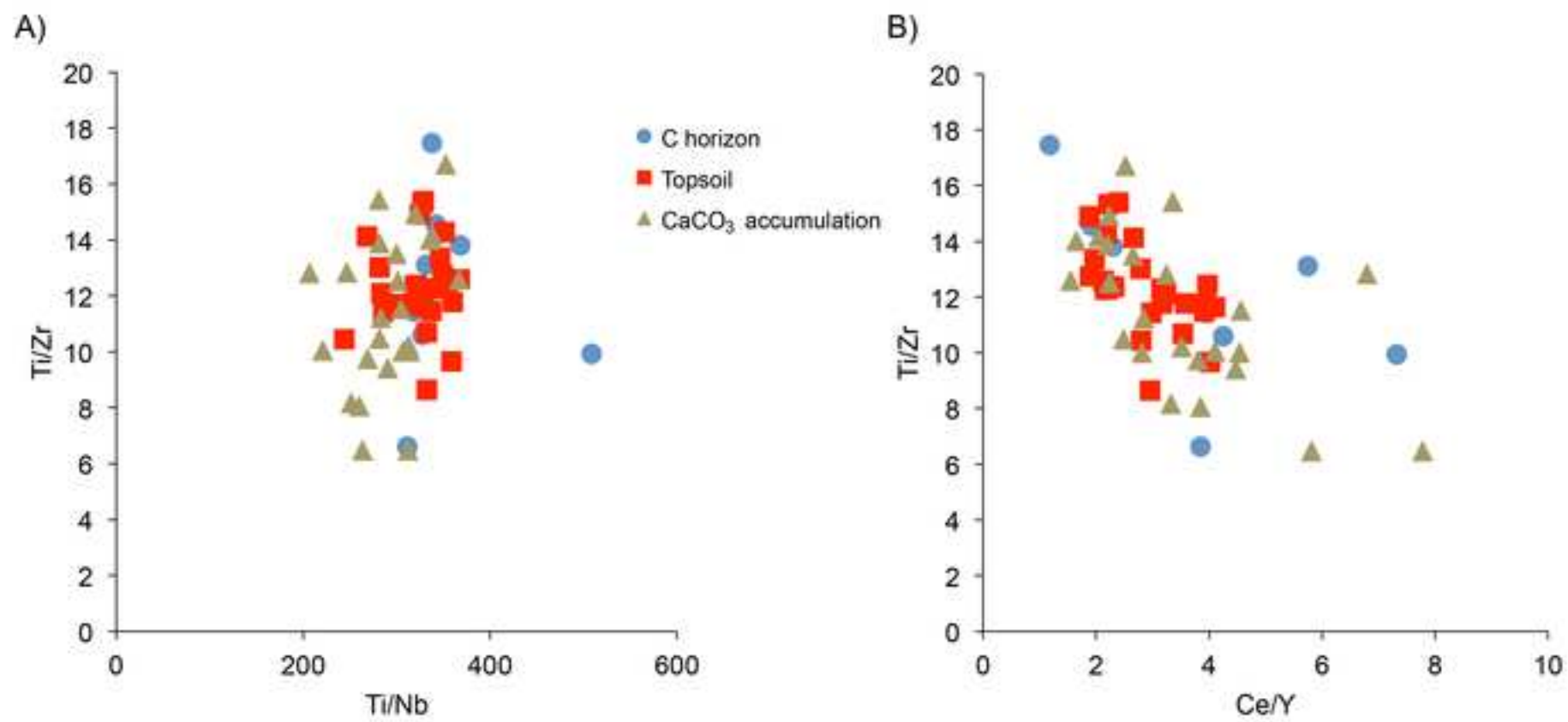


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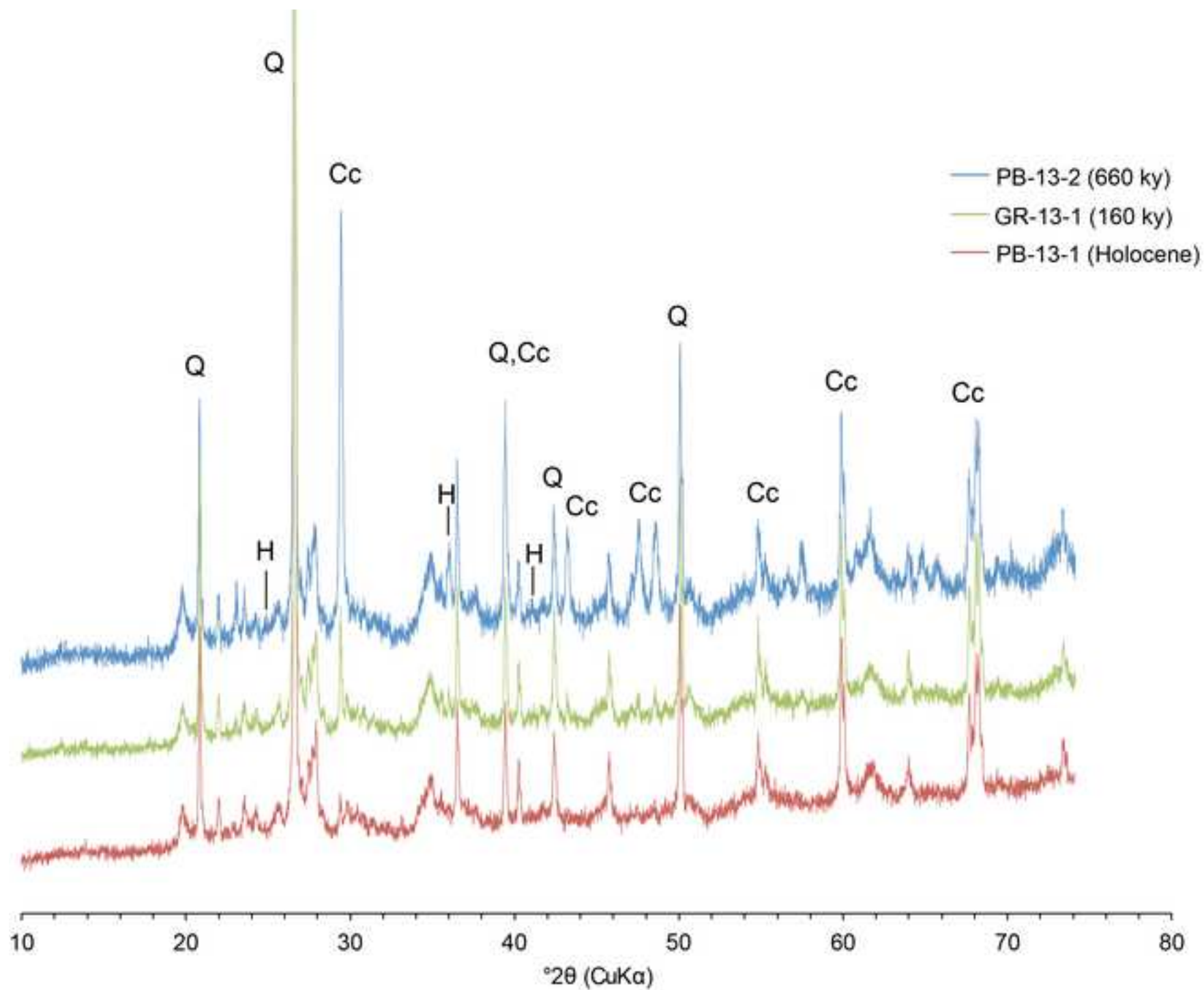


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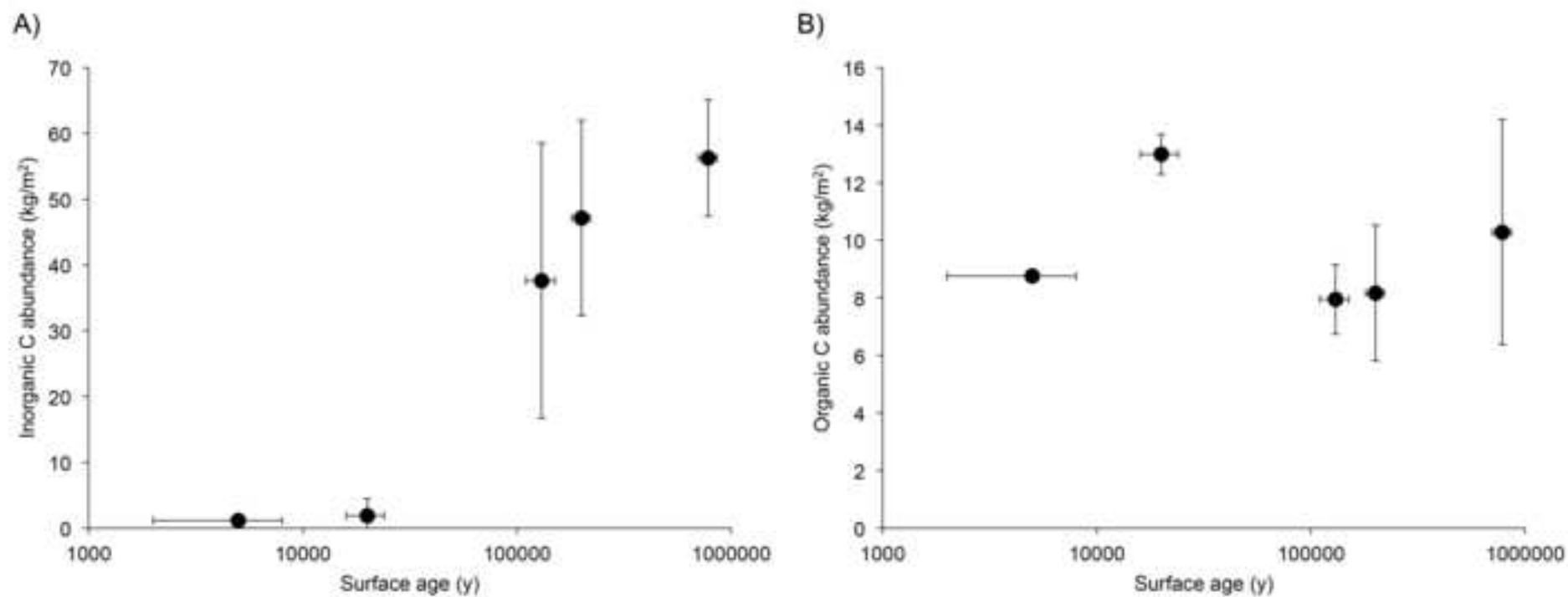


Figure 10

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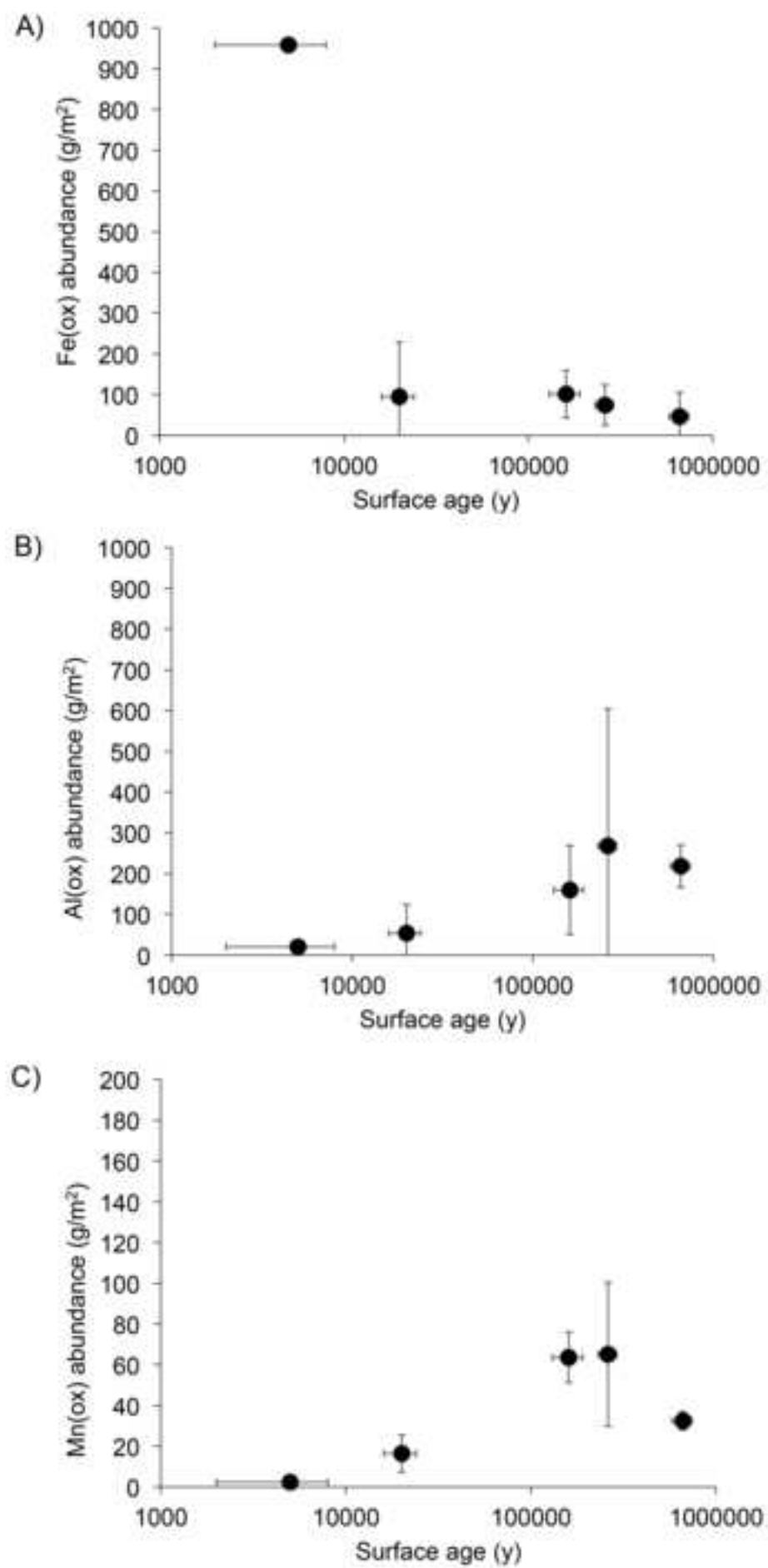


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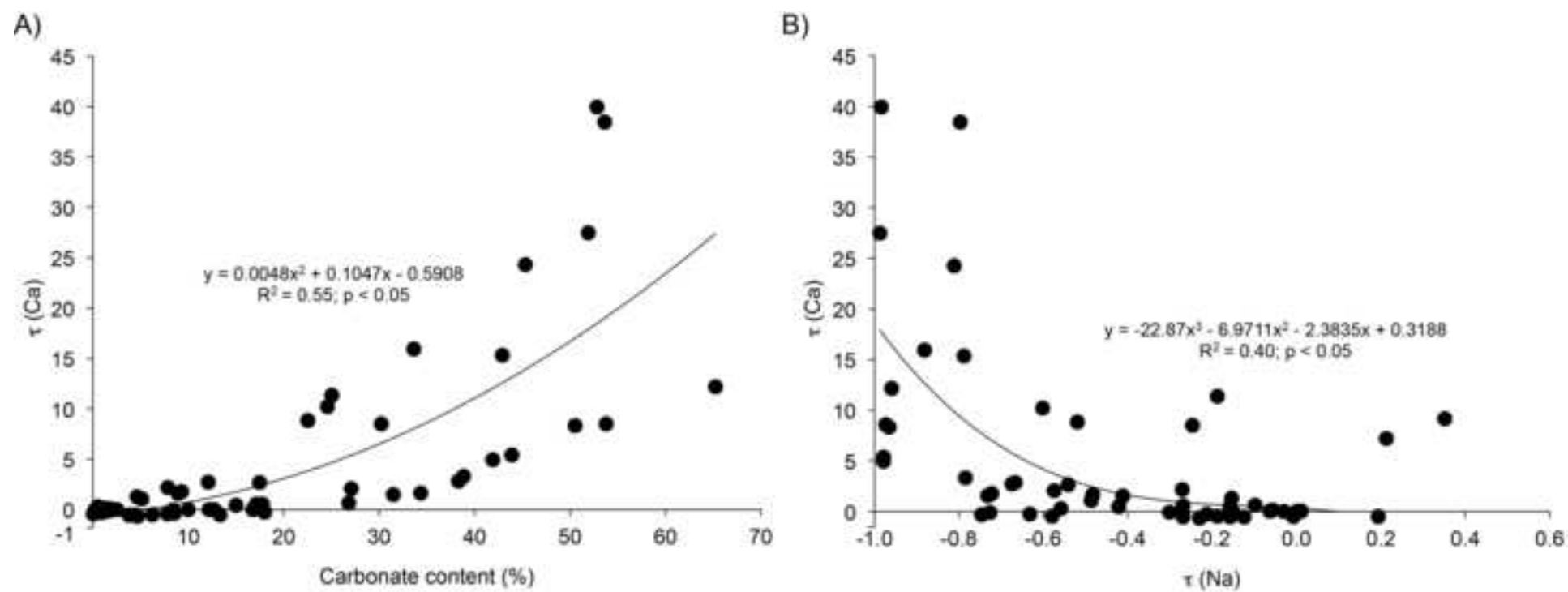


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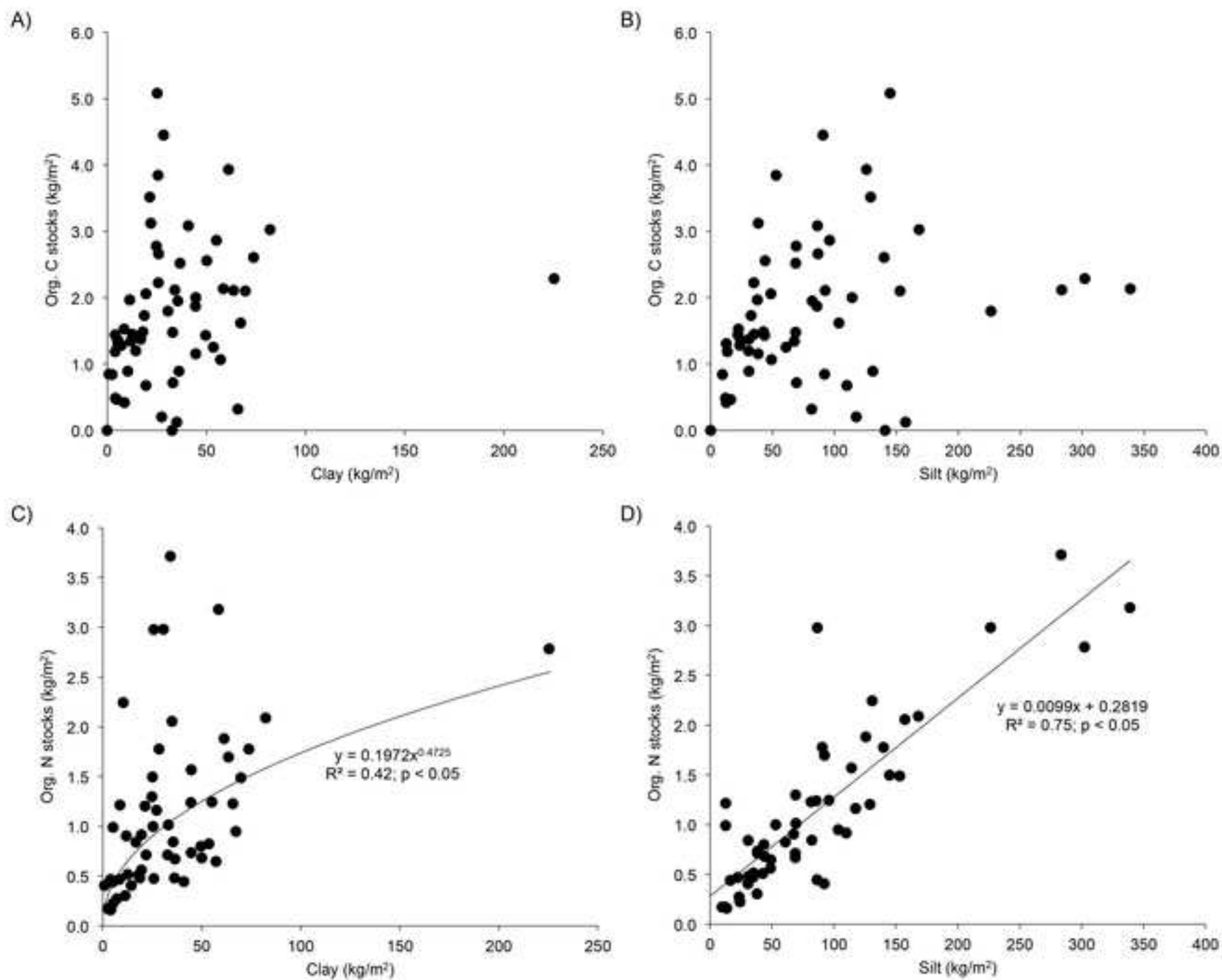


Table 1. Characteristics of the terrace sites along the North Fork, Middle Fork and Little Popo Agie rivers, Wind River Range.

Site / Soil profile	Longitude °E	Latitude °N	Elevation m asl	Parent Material*	Vegetation	Terrace age ¹⁾ ky	MAP ²⁾ mm/year	MAT ²⁾ °C
Little PopoAgie – Red Canyon								
RCT2	108°39'54"	42°40'39"	1722	RP/C	Grass, Sagebrush	20 (±4)	250-380	6.7
RCT3	108° 40'10"	42°40'40"	1750	RP/C	Grass, Sagebrush	160 (±30)	250-380	6.6
RCT4	108° 40'10"	42°40'24"	1768	RP/C	Grass, Sagebrush	260 (±30)	250-380	6.4
RCT5	108° 40'19"	42°40'17"	1795	RP/C	Grass, Sagebrush	660 (±40)	250-380	6.3
Middle Fork - Lander								
LWS-13-1	108° 43'34"	42°49'47"	1643	G	Grass, Sage, riparian	160 (±30)	330	7.2
SB	108°43'12"	42°49'30"	1676	G	Grass, Sagebrush	260 (±30)	330	7.0
North Fork								
PB-13-1	108°53'16"	42°52'08"	1826	OG	Grass, Sage, riparian	5 (±3)	250-380	6.1
GalPD	108°51'39"	42°53'03"	1768	RP/C	Grass, Sage, riparian	20 (±4)	250-380	6.4
GR-13-1	108°51'47"	42°53'12"	1798	G	Grass, Sagebrush	160 (±30)	250-380	6.3
GR-13-2	108°52'15"	42°52'46"	1838	G	Grass, Sagebrush	260 (±30)	250-380	6.0
PB-13-2	108°52'58"	42°52'28"	1908	G	Grass, Sagebrush	660 (±40)	250-380	5.6

¹⁾ Ages for the ~5, ~20, ~160 and ~260 ky terraces are assigned (a) according to their local morphostratigraphic positions and (b) their correlations to previously-dated terrace units along the Wind/Bighorn river (heights above the modern channels and associated incision rates (Chadwick et al., 1997; Dahms 2010; Hancock et al., 1999; Nettleton and Chadwick, 1991; Phillips et al., 1997; Reheis, 1987). The highest terraces (soils RCT5, PB-13-2) were assigned an age of ~660 ky according to their correlation to the ‘Airport’ terrace of the Middle Fork (‘AT’ of Fig. 2B), in which an ash deposit of Lava Creek age previously was reported (Anders et al., 2009; Jaworowski, 1992).

²⁾ Data source: Massatti (2007), Dahms et al. (2012), PRISM Climate Group (2014)

*G = granitic alluvium; RP/C = Red Peak/Chugwater fines over coarse granitic gravels; OG = organic-rich granitic.

Table 2. Physical characteristics of the terrace soils.

Locality/ profile	Horizon	Depth (cm)	Gravel w.-%	Gravel vol.-%	Particle Size Distribution			Bulk density (g/cm ³)
					Sand (2-0.5 mm)	Silt (500-2μ)	Clay (<2μ)	
Little Popo Agie - Red Canyon RCT2	A	0-9	0.6	0.3	27.2	49.2	23.6	1.2
	Bw1	9-21	0.9	0.5	22.1	51.3	26.6	1.4
	Bw2k	21-40	0.1	0.1	25.0	49.1	25.9	1.5
	CBk	40-60	0.2	0.2	26.4	49.5	24.2	1.7
	CK	60-90+	1.0	0.7	25.9	63.2	10.9	1.8
RCT3	A	0-7	14.2	7.0	39.1	44.9	16.1	1.2
	AB	7-30	13.2	7.4	37.3	42.2	20.5	1.4
	Bk1	30-50	22.3	14.0	38.5	44.2	17.3	1.5
	Bk2	50-100	32.1	23.3	60.6	34.7	4.7	1.7
	2C	100+	25.8	19.1	55.0	36.0	8.0	1.8
RCT4	A	0-10	23.3	12.1	35.5	46.1	18.5	1.2
	Bw1	10-18	14.7	8.4	40.9	41.6	17.5	1.4
	Bw2	18-30	34.2	22.8	27.0	49.4	23.6	1.5
	Bk1	30-50	33.9	21.3	22.5	47.0	30.5	1.4
	Bk2	50-100	14.6	9.4	27.2	41.7	31.1	1.6
	Ck	100-125	54.3	44.7	48.1	44.2	7.8	1.8
	C	125+	49.4	39.8	55.4	36.2	8.4	1.8
RCT5	A	0-15	6.6	3.1	32.4	47.1	20.4	1.2
	AB	15-25	30.6	17.8	52.1	30.5	17.4	1.3
	Bk	25-40	68.4	56.6	11.0	88.3	0.8	1.6
	Bkm	40-80	87.7	81.1	--	--	--	1.6
	Ck	80-200	55.4	45.8	90.3	7.4	2.2	1.8

Middle PopoAgie

- Lander LWS-13-1	A	0-13	12.8	6.2	35.8	30.0	34.2	1.2
	Bt1	13-23	3.8	2.2	27.6	33.4	39.0	1.5
	Bt2	23-32	10.4	6.2	47.1	24.4	28.5	1.5
	Bk	32-120	37.0	28.5	71.9	25.0	3.0	1.8
	BCK	120-150	27.2	18.4	62.4	20.8	16.8	1.6
	C	150+	36.4	28.0	55.4	36.2	8.4	1.8
SB	A	0-10	14.9	7.3	59.9	27.4	12.8	1.2
	Bt	10-25	6.3	3.7	47.2	28.1	24.7	1.5
	Bk1	25-60	23.0	13.6	63.2	21.8	15.0	1.4
	Bk2	60-100	20.9	12.2	71.3	26.6	2.1	1.4
	C	100+	36.4	28.0	55.4	36.2	8.4	1.8
North Fork PopoAgie PB-13-1	A	0-5	8.6	4.1	70.0	23.4	6.6	1.2
	AB	5-18	10.3	5.3	69.3	23.6	7.1	1.3
	B	18-50	15.7	9.6	70.7	19.9	9.4	1.5
	BC	50+	15.9	9.7	74.1	16.9	9.0	1.5
GalPD	A	0-25	1.0	0.4	31.8	58.1	10.1	1.0
	Bw	25-45	0.4	0.2	31.5	58.8	9.7	1.1
	BC	45-70	2.7	1.2	24.9	51.6	23.5	1.2
	C1	70-90		0.0	55.2	30.3	14.5	1.2
	C2	90-120						
	C3	120+						
GR-13-1	Ak	0-10	25.7	11.5	70.7	24.9	4.4	1.0
	Btk	10-21	42.9	25.4	68.7	22.7	8.5	1.2
	Bk	21-52	43.5	31.7	72.2	20.4	7.3	1.6
	BCK	52-75	53.1	37.4	89.5	8.1	2.4	1.4
	Ck	75+	37.8	25.6	85.9	9.2	5.0	1.5
GR-13-2	A	0-5	8.5	3.4	66.4	25.1	8.5	1.0

PB-13-2	Bt1	5-18	12.3	8.7	56.5	20.3	23.2	1.8
	Bt2	18-30	13.9	9.8	57.5	19.7	22.9	1.8
	Bk	30-70	41.2	27.0	73.7	20.1	6.3	1.4
	C2	70-90	38.6	13.1	92.9	5.0	2.1	0.6
	C3	90+	36.4	12.1	94.6	3.2	2.2	0.6
	A	0-5	9.7	3.2	68.8	24.9	6.3	0.8
	Bt1	5-13	14.4	7.1	65.6	26.4	7.9	1.2
	Bt2	13-20	15.4	9.3	69.1	25.4	5.4	1.5
	Bt3k	20-30	22.5	16.5	59.8	23.1	17.1	1.8
	Bk1	30-50	43.8	37.0	76.1	15.2	8.7	2.0
	Bk2	50-90	40.3	33.8	71.5	18.1	10.4	2.0
	BCK / CBk	90+	27.9	19.0	84.7	13.0	2.3	1.6

Table 3
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Table 3. Typical chemical characteristics of the terrace soils.

Locality	Horizon	Depth cm	pH (CaCl ₂)	inorg. C g/kg	org. C g/kg	org. N g/kg	Org. C/N	Fe (ox) ¹⁾ mg/kg	Al (ox) ¹⁾ mg/kg	Mn (ox) ¹⁾ mg/kg
Little PopoAgie - Red Canyon										
RCT2	A	0-9	7.31	5.0	35.7	9.26	3.9	350	647	158
	Bw1	9-21	7.44	5.7	11.2	7.43	1.5	367	726	168
	Bw2k	21-40	7.39	5.9	9.2	6.24	1.5	327	689	172
	CBk	40-60	7.58	10.6	8.9	6.15	1.4	305	666	214
	CK	60-90+	7.68	21.3	4.0	5.93	0.7	316	612	158
RCT3	A	0-7	7.52	6.3	18.5	6.62	2.8	223	589	323
	AB	7-30	7.58	10.9	13.2	6.32	2.1	182	529	264
	Bk1	30-50	7.67	50.9	7.8	6.08	1.3	24	116	58
	Bk2	50-100	7.92	49.0	2.8	4.57	0.6	76	207	92
	2C	100+	7.93	35.3	0.3	4.70	0.1	270	10	186
RCT4	A	0-10	7.39	10.1	19.5	5.32	3.7	375	1026	284
	Bw1	10-18	7.53	18.6	14.5	4.97	2.9	227	774	174
	Bw2	18-30	7.58	32.9	10.6	5.12	2.1	99	650	93
	Bk1	30-50	7.62	53.2	7.3	4.32	1.7	53	451	59
	Bk2	50-100	7.78	65.3	3.2	3.84	0.8	11	150	42
	Ck	100-125	8.03	48.3	2.7	3.68	0.7	120	283	105
	C	125+	7.80	22.0	0.6	3.58	0.2	70	43	265
RCT5	A	0-15	7.64	23.4	11.2	4.84	2.3	202	534	223
	AB	15-25	7.57	39.3	16.2	4.49	3.6	123	336	107
	Bk	25-40	7.73	62.6	8.1	3.92	2.1	45	104	35
	Bkm	40-80	7.91	81.7	2.7	3.09	0.9	19	10	48
	Ck	80~200	7.99	43.8	2.3	2.54	0.9	91	130	132

Middle

PopoAgie -
Lander
LWS-13-1

A	0-13	7.70	14.9	17.5	4.67	3.7	523	440	179
Bt1	13-23	7.69	11.1	7.3	4.42	1.7	413	438	214
Bt2	23-32	7.71	11.5	7.1	3.82	1.8	172	665	134
Bk	32-120	7.70	40.5	1.9	3.28	0.6	10	72	37
BCk	120-150	7.89	22.4	0.8	3.14	0.3	52	91	127
C	150+								

SB

A	0-10	7.37	0.7	10.8	3.66	2.9	285	347	206
Bt	10-25	7.49	0.9	5.8	3.80	1.5	364	491	234
Bk1	25-60	7.69	63.3	5.0	4.01	1.2	47	131	20
Bk2	60-100	7.88	53.0	1.8	4.57	0.4	79	78	37
C	100+								

North Fork
PopoAgie
PB-13-1

A	0-5	6.40	0.6	20.6	2.82	7.3	1120	353	193
AB	5-18	6.15	0.4	12.3	1.91	6.4	2267	335	260
B	18-50	6.80	0.1	7.1	1.02	7.0	770	418	190
BC	50+	7.05	2.5	6.2	1.65	3.8	2557	435	185

GalPD

A	0-25	7.50	8.3	20.4	6.01	3.4	400	558	155
Bw	25-45	7.60	17.5	16.0	5.47	2.9	392	623	188
BC	45-70	7.70	22.1	7.1	5.02	1.4	218	236	211
C	70-90+	7.78	16.1	3.1	4.45	0.7	83	331	163
	90-120	7.83	15.4	1.5	3.47	0.4	10	241	337
	>120	7.80	12.9	1.5	3.96	0.4	10	164	506

GR-13-1

Ak	0-10	7.30	0.4	16.3	5.26	3.1	591	805	273
Btk	10-21	7.35	0.1	15.5	4.75	3.3	258	1073	219
Bk	21-52	7.60	28.1	8.2	3.83	2.1	165	434	111
BCk	52-75	7.50	18.6	2.3	2.19	1.0	166	100	80
Ck	75+	7.30	15.3	4.1	2.51	1.6	168	100	73

GR-13-2	A	0-5	6.18	0.0	10.1	3.57	2.8	385	287	149
	Bt1	5-18	6.53	1.4	6.7	3.74	1.8	271	893	100
	Bt2	18-30	6.80	2.3	5.9	3.77	1.6	130	1073	45
	Bk	30-70	7.54	29.5	9.9	3.95	2.5	48	669	3
	C2	70-90	7.74	54.4	5.2	3.93	1.3	107	33	7
	C3	90+	7.72	6.5	1.1	3.12	0.3	581	116	29
PB-13-2	A	0-5	7.50	1.4	21.7	4.46	4.9	350	505	251
	Bt1	5-13	7.30	1.0	14.4	3.09	4.7	291	514	229
	Bt2	13-20	7.50	7.0	14.3	2.38	6.0	180	568	162
	Bt3k	20-30	7.65	19.8	14.8	3.15	4.7	138	412	96
	Bk1	30-50	7.60	58.1	12.4	2.82	4.4	25	46	26
	Bk2	50-90	7.20	56.4	5.4	2.35	2.3	24	40	12
	BCk / CBk	90+	7.65	22.6	2.6	1.75	1.5	114	40	46

¹⁾ox = oxalate-extractable fraction

Table 4. Overview of the main soil minerals (obtained from XRD and DRIFT measurements) in the fraction < 32 µm (clay + fine silt)

Site (age)	Horizon	Quartz	Orthoclase	Albite	Mica	Vermiculite	Chlorite	Calcite	Dolomite	Kaolinite	Hematite
PB-13-1 (Holocene)	A	xxx	xx	xx	x				?	x	?
	AB	xxx	x	x	x	x	?	?	?	x	?
GR-13-1 (160 ky)	Ak	xxx	x	x	x	x		xx	x	x	(x)
	Btk	xxx	x	x	x	x		xx	x	x	(x)
	Bk	xxx	x	x	x	x		xx	?		
	Ck	xx	x	x	x	?		xxx	xx	x	?
PB-13-2 (660 ky)	A	xxx	xx	xx	x	x		x	?	x	(x)
	Bt1	xxx	xx	xx	xx	x		xx	?	x	(x)
	Bt2	xxx	x	xx	x	x		xxx	x	x	(x)
	Bt3k	xxx	x	x	x	?		xxx	?	x	(x)
	Bk1	xx	x	x	x	?		xxx	x	x	?
	BCK/CBk	xx	x	x	?	x		xxx	xx	(x)	(x)

Concentration indications
? questionable
(x) traces
x low amount
xx moderate amount
xxx high amount

Table 5. Stocks and interval-accumulation rates for pedogenic CaCO₃.

Terrace age	CaCO ₃ stocks kg/m ²	Average accumulation rates (linear trend) g/m ² /y	Time range (for accumulation rate)	Interval- Accumulation rates g/m ² /y
20 ky	16	0.79	0 - 20 ky	0.79
160 ky	314	1.96	20 - 160 ky	2.13
260 ky	393	1.51	160 - 260 ky	0.79
660 ky	469	0.71	260 - 660 ky	0.19

Appendix A

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